

**National Low-Level Waste
Management Program
Radionuclide Report Series
Volume 17: Plutonium-239**

Published March 1999

**National Low-Level Waste Management Program
Radionuclide Report Series**

Volume 17: Plutonium-239

**J. P. Adams
M. L. Carboneau**

Published March 1999

**Idaho National Engineering and Environmental Laboratory
National Low-Level Waste Management Program
Lockheed Martin Idaho Technologies Company
Idaho Falls, Idaho 83415**

**Prepared for the
U.S. Department of Energy
Office of Environmental Restoration and Waste Management
Under DOE Idaho Operations Office
Contract DE-AC07-94ID13223**

ABSTRACT

This report, Volume 17 of the National Low-Level Waste Management Program Radionuclide Report Series, discusses the radiological and chemical characteristics of plutonium-239 (^{239}Pu). This report also discusses waste types and forms in which ^{239}Pu can be found, waste and disposal information on ^{239}Pu , and ^{239}Pu behavior in the environment and in the human body.

FOREWORD

The purpose of the National Low-Level Waste Management Program Radionuclide Report Series is to provide information to state representatives and developers of low-level radioactive waste disposal facilities about the radiological and chemical characteristics of selected radionuclides and their behavior in the low-level radioactive waste disposal facility environment. Extensive surveys of available literature provided information that was used to produce this series of reports and an introductory report.

The National Low-Level Waste Management Program Radionuclide Report Series previously addressed the following radionuclides: technetium-99, carbon-14, iodine-129, tritium, cesium-137, strontium-90, nickel-59, plutonium-241, nickel-63, niobium-94, cobalt-60, curium-242, americium-241, uranium-238, and neptunium-237. These radionuclides usually offer the greatest contribution to the dose estimated during a performance assessment analysis.

This report is Volume 17 of the series. It outlines the basic radiological and chemical characteristics of plutonium-239, waste types and forms that contain it, waste and disposal information, and its behavior in environmental media such as soils, plants, groundwater, air, and in the human body.

CONTENTS

ABSTRACT	iii
FOREWORD	v
INTRODUCTION	17-1
RADIOLOGICAL CHARACTERISTICS	17-2
CHEMICAL AND PHYSICAL CHARACTERISTICS	17-9
PLUTONIUM-239 PRODUCTION IN NUCLEAR REACTORS	17-11
WASTE AND DISPOSAL DATA ON PLUTONIUM-239	17-14
Nuclear Reactors	17-14
Medical, Academic Institutions, and Industrial Plutonium-239 Wastes	17-14
Disposal Data on Plutonium-239	17-15
BEHAVIOR OF PLUTONIUM IN THE ENVIRONMENT	17-19
Plutonium in Soils	17-19
Plutonium in Water	17-22
Plutonium in Plants	17-23
Plutonium in Air	17-24
BEHAVIOR OF PLUTONIUM IN THE HUMAN BODY AND IN ANIMALS	17-26
SUMMARY	17-30
REFERENCES	17-31
BIBLIOGRAPHY	17-37

FIGURES

17-1. Fission product yield curve for thermal fission of ^{235}U and ^{239}Pu . The curve with diamonds represents ^{235}U , and the curve labeled with boxes represents ^{239}Pu	17-13
17-2. Pu-239 activity deposited at the Richland, Beatty, and Barnwell sites by year	17-16
17-3. Pu-239 activity deposited at the Richland, Beatty, and Barnwell sites by source.	17-17
17-4. Pu-239 activity deposited at the Richland and Beatty sites by waste type.	17-18

TABLES

17-1. The primary α -particle energies released from ^{239}Pu decay with subsequent γ -rays released from the excited state ^{235}U daughter nucleus.	17-3
17-2. A mass-energy balance for Pu-239 decay resulting in the emission of a 5.105-MeV α -particle and a 52-keV gamma-ray.	17-6
17-3. Comparison of the average and maximum kinetic energies of atomic electrons (e^-), beta-particles (β), and α -particles released during the decay of several radionuclides	17-7
17-4. Comparison of the radiotoxicity of several important radionuclides including ^{239}Pu	17-8
17-5. Physical properties of plutonium.	17-10
17-6. Plutonium-239, total plutonium, total actinide, and total fission product inventories calculated as a function of decay time following a 3-year irradiation in a 3,200-MW thermal power LWR producing a total fuel burnup of 33,400 MWtD/MTU.	17-12
17-7. Annual limits on intake (ALI) and the derived air concentrations (DAC) for ^{239}Pu for ingestion and inhalation.	17-28

National Low-Level Waste Management Program

Radionuclide Report Series

Volume 17: Plutonium-239

INTRODUCTION

This report describes the basic radiological and chemical characteristics of plutonium-239 (^{239}Pu) and examines how these characteristics affect the behavior of ^{239}Pu in various environmental media such as soils, groundwater, plants, the atmosphere, and the human body. Discussions also include methods of ^{239}Pu production, radiation, waste types and forms that contain ^{239}Pu , and waste disposition information on ^{239}Pu . In general, the International System of Units (SI) is used throughout this report. However, some historical (non-SI) units are used in many of the references and it was decided to preserve these units in some cases. The SI unit for radioactivity is the Becquerel (Bq), which is one disintegration per second, whereas the historical unit for radioactivity is the Curie (Ci), which is 3.7×10^{10} disintegrations per second - therefore, $1 \text{ Ci} = 3.7 \times 10^{10} \text{ Bq}$. The SI unit for dose (the biological effects of radiation on humans) is the Sievert (Sv). The historical unit for dose is the Roentgen Equivalent Man, or rem. The relation between the two dose units is $1 \text{ Sv} = 100 \text{ rem}$.

All plutonium atoms contain 94 protons ($Z=94$) and various numbers of neutrons (e.g., $N=138$ to 152) within the atom's nucleus. At least fifteen isotopes of plutonium are known to exist (namely, ^{232}Pu through ^{246}Pu).¹ All isotopes of plutonium are radioactive. Except for trace quantities of plutonium in pitchblende (e.g., one part ^{239}Pu in 10^{11} atoms),² and small amounts produced in natural reactors,^a ^{239}Pu does not normally exist as a naturally occurring nuclide. Although ^{239}Pu is not a normal component of the natural background activity, small quantities of it have been released into the environment, primarily from plutonium weapons testing and nuclear reactor accidents.

^{239}Pu is the most common plutonium isotope, with ^{240}Pu representing the second most common form. ^{239}Pu is a fissile material, meaning that it will fission with slow or thermalized neutrons. Small quantities of PuO_2 (enriched to $>99\%$ ^{239}Pu) can be purchased from the Isotope Distribution Office of the Oak Ridge National Laboratory (ORNL) at a cost of \$1.25 per mg ($>99\%$ ^{239}Pu) to \$4.80 per mg (at 99.99% ^{239}Pu) plus \$2,000 to \$3,000 for the container plus a shipping charge.³ Since plutonium is classified as a special nuclear material, its purchase requires a DOE/NRC (Department of Energy/Nuclear Regulatory Commission) transfer accountability form with an NRC or State agreement license.

^{239}Pu is present in all plutonium-based nuclear weapons to an extent of approximately 93% of the total plutonium mass, with the exact enrichment quantity being a classified value.⁴ During the course of nuclear weapons testing from 1945 to 1976, approximately 360,000 to 500,000 Ci of plutonium isotopes (estimated ^{239}Pu mass of $>2,200 \text{ kg}$ with some estimates indicating as much as 5 tons of plutonium⁵) have been deposited in the Earth's biosphere; much of it is contained in the Northern Hemisphere.^{5,6,7,8} The plutonium that was originally released into the atmosphere has eventually settled out onto land or ocean regions. That which has been deposited on the land now exists within the top 10 cm of soil, and 50% of the plutonium that was deposited within the Earth's oceans is now contained below the thermocline (defined as a layer in a thermally stratified body of water that separates an upper, warmer, lighter, oxygen-rich zone from a lower, colder, heavier, oxygen-poor zone).⁵

a. For example, the Oklo natural reactor in Gabon produced several tons of ^{239}Pu during the reactor's active lifetime of $\sim 500,000$ years approximately 2 billion years ago.

Additional quantities of ^{239}Pu have been released to the environment from nuclear reactor accidents (e.g., the Windscale accident⁶ in 1957 and the Chernobyl reactor accident⁹ in 1986); from nuclear powered submarine accidents (e.g., U.S. Navy submarines Thresher and Scorpion accidents in 1963 and 1968 plus USSR Navy submarines at various dates); an aerial refueling explosion involving a B-52 bomber carrying four plutonium based nuclear weapons in 1966 near the Spanish village of Palomares, Spain; and a B-52 crash near Thule Air Force Base in 1968).⁶⁻⁸ Other accidents have also released small amounts of plutonium to the environment.^{10,5} However, the total amount of the accidental releases of plutonium have been relatively minor on a global scale compared with the intentional releases from weapons testing; although, the local effects of these accidental releases are sometimes significant. In comparison with the above accidental releases of ^{239}Pu , the Savannah River nuclear fuel reprocessing facilities release less than 0.001 Ci (0.016 g) of ^{239}Pu in particulate form to the atmosphere per year.¹⁰

RADIOLOGICAL CHARACTERISTICS

^{239}Pu has a specific activity of 0.062 Ci/g (0.062 Ci radioactivity per gram of ^{239}Pu), and has a relatively long half-life ($T_{1/2}$) of 24,100 years.¹ ^{239}Pu can be produced from ^{238}U in any nuclear reactor from either thermal, intermediate, or fast energy neutrons. The specific nuclear reaction that produces ^{239}Pu is neutron capture (n,γ) in ^{238}U with subsequent beta-decay (i.e., electron emission from the radioactive nucleus) as follows: $^{238}\text{U} + n \rightarrow ^{239}\text{U} \xrightarrow{\beta,\gamma} ^{239}\text{Np} \xrightarrow{\beta,\gamma} ^{239}\text{Pu}$. Once formed, ^{239}Pu undergoes radioactive decay by emitting one of several possible alpha-particles (α -particles) to form ^{235}U . The three most likely released α -particle energies, and the corresponding branching ratios are: 5.156 MeV (73.3%), 5.144 MeV (15.1%), and 5.105 MeV (11.5%).^{11,12,13} In addition to the above three α -particles, at least 47 additional low-probability α -particle emissions of energies ranging from 4.06 MeV to 5.1 MeV are known; however, the total emission probability for these events is <0.2%.¹² The average α -particle energy from ^{239}Pu decay is ~5.15 MeV.⁵ The average distance in air that a 5.2 MeV α -particle can travel is about 4 cm (at 15°C and 1 atmosphere pressure).¹⁴ Also, it requires an α -particle of at least 7.5 MeV of energy to penetrate the protective layer of human skin (0.07 mm thick); therefore, the α -particles from ^{239}Pu decay cannot penetrate the outer layers of the epithelium.^{15,5} In fact, the range of these α -particles within bone and human tissue are respectively: 0.024 mm and 0.04 mm.⁵ Table 1 shows a more detailed listing of the release probabilities and energies associated with α -particles produced from ^{239}Pu decay, and the corresponding gamma-ray energies released from the excited state ^{235}U daughter nucleus.

The decay of ^{239}Pu does not result in the release of β -particles nor high-energy γ -rays. Nevertheless, low-energy γ -rays (e.g., photons released from the atom's nucleus) are released from the excited state ^{235}U daughter nucleus following alpha emission from the ^{239}Pu parent nucleus. In addition, two atomic (orbital) electrons from ^{239}Pu are also lost after the emission of the α -particle. The γ -ray photons that are released from the ^{235}U daughter nucleus are usually called uranium x-rays because of the magnitude of their energies. The most likely photon energies are: 0.0130 MeV, 0.0386 MeV, 0.0516 MeV; however, many others are possible (see Table 1). A large number of low-energy γ -rays are produced from the excited ^{235}U nucleus, with 99.9% having energies less than 0.100 MeV (100 keV). The average photon energy is about 0.010 MeV (~10 keV). In fact, the radiological dose received by a person positioned 2 feet away from an unshielded 1,000-g spherical mass of metallic ^{239}Pu would be ~0.04 mrem/hr.¹⁶ In comparison, an average person in the U.S. receives a dose of ~0.023 mrem/hr (or ~200 mrem/year) from radon gas, and a total dose of 360 mrem per year from all sources.¹⁷ The external dose received from ^{239}Pu is due primarily to γ -rays, with the released α -particles and electrons contributing almost nothing to the total dose.

^{239}Pu can also decay via spontaneous fission; however, the half-life of this disintegration event is 5.5×10^{15} years.¹¹ The total amount of heat generated within a 1,000-g (1-kg) mass of ^{239}Pu is 1.92 watts,

with ~99.9% of this heat produced by the deposition of particle kinetic energy (mainly α -particles) and the remainder being gamma-ray heat. In any sizeable quantity of plutonium metal, essentially all of the α -particle energy and a large fraction of the γ -ray energy will be deposited within the metal in the form of heat.

Table 17-1. The primary α -particle energies released from ^{239}Pu decay with subsequent γ -rays released from the excited state ^{235}U daughter nucleus. Many additional α -particle and γ -ray energies are known but are not shown in this table. Information obtained from References 12 and 13.

α -Particle Release Probability From ^{239}Pu	α -Particle Energy From ^{239}Pu Decay (MeV)	γ -ray Release Probabilities and Photon Energy From $^{235}\text{U}^a$ (MeV)	Total $\alpha+\gamma$ Energy (MeV)
0.0180%	5.008	73% 0.098 + 0.0516 27% 0.098 + 0.0386 + 0.0130	5.158
0.005%	5.029	77% 0.129 14% 0.116 + 0.0130 6.6% 0.0776 + 0.0516 2.4% 0.0776 + 0.0386 + 0.0130	5.158
0.030%	5.055	20% 0.1030 80% 0.0568 + 0.0462	5.158
0.036%	5.076	100% 0.0686 + 0.0130	5.158
11.5%	5.105	73% 0.0516 27% 0.0386 + 0.0130	5.157
0.015%	5.111	100% 0.0462	5.157
15.1%	5.144	100% 0.0130	5.157
73.3%	5.156	100% 0.00008	5.156
Total = ~100%	Average ^b α -particle energy per ^{239}Pu decay is 5.148	Average ^b γ -ray energy per ^{239}Pu decay is ~0.010	~5.158

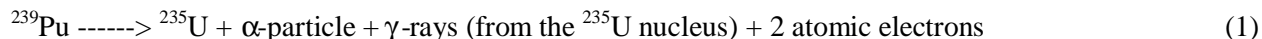
a. ^{235}U is the daughter atom that is produced following ^{239}Pu decay. The γ -ray photon(s) are actually released from the excited state ^{235}U nucleus following ^{239}Pu decay, and not from the ^{239}Pu nucleus.

b. The reported average energies represent estimated mean values computed over all α -particle energies or all released γ -ray energies, not just those listed in this table. An average α -particle energy of 5.15 MeV is reported in Reference 5.

The decay scheme for ^{239}Pu , showing the emission of a single α -particle (i.e., ^4He nucleus with no orbital electrons) is shown in Equation (1). Also shown in Equation (1) is the emission of two atomic electrons, which will be explained later. The transition from an excited-state $^{235}\text{U}^*$ nucleus to the ground-state ^{235}U nucleus is very short and is neglected in Equation (1). A neutrally charged ^{239}Pu atom contains 94 protons (within the atom's nucleus) with 94 electrons orbiting the positively charged nucleus. The neutrally charged ^{235}U daughter atom contains only 92 protons (within the atom's nucleus) and 92 electrons orbiting the positively charged nucleus. Therefore, there are 2 electrons not needed by the newly formed uranium atom, and can be released shortly after the α -particle emission. This will eventually form a neutrally charged ^{235}U atom. In this case, the α -particle represents an ionized helium atom (e.g., $^4\text{He}^{+2}$) without any orbital electrons. Immediately after the α -particle is released from the ^{239}Pu nucleus, it traverses the electron cloud around the newly formed daughter atom ($^{235}\text{U}^{++}$), imparting some of its kinetic energy to the orbital electrons. The mass-energy and particle emission events corresponding to ^{239}Pu decay are shown in Table 2. The data in Table 2 represent only one of many

possible mass-energy particle emission scenarios whereby the released energy from ^{239}Pu decay is distributed between an α -particle, a γ -ray, two lost atomic electrons, and the recoil kinetic energy of the ^{235}U atom.

PU-239 DECAY TRANSFORMATION:



As mentioned before, several α -particle and γ -ray energies are possible in Equation (1). The most prominent α -particle and γ -ray energies are shown in Table 1. Table 2 presents a detailed illustration of Equation (1) showing a mass-energy balance for a specific decay scenario, resulting in the emission of a 5.105 MeV α -particle and a 52 keV γ -ray. Instead of a single 0.052 MeV (52 keV) γ -ray as shown in Table 2, it is possible that two lower energy γ -rays (e.g., 13 keV and 39 keV) can be released for this particular decay scenario. In any event, the mass-energy balance shown in Table 2 represents only one particular situation leading to the release of an α -particle. Notice that the released atomic electrons for this decay scenario are not very energetic.

For the decay scenario illustrated in Table 2, the total kinetic energy of the released electrons is only 30 keV. This represents a nominal situation; however, there are some very rare events whereby the released electrons can be very energetic (See Reference 12). Electrons with energies of only 30 keV will have very limited penetrating capabilities. For example, electrons with energies of ~20 keV have a maximum range of less than 20 mm in air and less than 0.05 mm in water. Also, to penetrate the protective layer of human skin, an electron would have to have an energy of at least 70 keV.¹⁵ Therefore, the α -particles and electrons released from ^{239}Pu decay are not directly hazardous when the source of plutonium is outside the body. The general perception that “plutonium is the most toxic substance known to humans” is an exaggeration of the truth.⁵ In fact, ^{239}Pu is only hazardous to humans in certain forms and if it is acquired through specific pathways (e.g., the lungs). The direct irradiation effects of ^{239}Pu are actually quite minimal.

As shown in Column 3 of Table 2, the total decay energy associated with ^{239}Pu decay is ~5.27 MeV, of which the α -particles carry off ~97%, the recoiling daughter nucleus carries off 1.6%, the γ -ray(s) <1%, and the released (atomic) electrons only ~0.5% of the available energy.

Table 3 compares the average and maximum energy of electrons and α -particles released from several different radioactive isotopes. The released electrons can be generated from either beta-particles ($\beta = e^-$), released from the nucleus of the atom (e.g., $n \rightarrow p^+ + e^-$), or from the loss of atomic (orbital) electrons after the initial decay event due to many different reasons. The electron release from ^{239}Pu decay is actually the result of too many atomic electrons surrounding the ^{235}U nucleus. A neutrally charged plutonium atom has 94 protons and 94 electrons; however, after α -particle decay, the plutonium nucleus has produced a uranium nucleus that contains 92 protons but also includes the original 94 orbital electrons (two more than is needed to balance the nuclear charge). Usually the extra electrons will be lost shortly after the uranium atom is created. Therefore, the electrons released from ^{239}Pu decay are the result of atomic forces occurring in the ^{235}U atom and not from the ^{239}Pu nucleus; however, this distinction is only technical since all electrons (regardless of how they are produced) are identical. From the data shown in Table 3, it is evident that the average electron energy from ^{239}Pu decay is small compared with other radionuclides. In fact, it requires a 70-keV electron to penetrate the protective layer of human skin. Also, the maximum range in air of most electrons (>99%) from ^{239}Pu decay is less than 6 inches. Compared to other α -emitters, the α -particle energies from ^{239}Pu are fairly typical.

Table 4 lists the radiotoxicity of several important radionuclides. Both ^{239}Pu and ^{241}Pu appear in the same very high radiotoxicity (Group 1) classification. This is also true of the other isotopes of plutonium. The principal reason that ^{239}Pu is listed in the highest radiotoxicity group is that it emits very-

high-energy α -particles. Alpha-particles are generally less hazardous than other forms of radiation outside the human body because they are stopped by very thin materials such as skin. Thus, ^{239}Pu does not pose a significant external radiation hazard. However, if ^{239}Pu enters into the human body (by inhalation, ingestion, or other means), it can pose a significant radiation hazard because of the high energy associated with its alpha emissions. Alpha-particles deposit all of their energy within a thin layer of living tissue, for example in the lung, resulting in an increased chance of cell damage. Since ^{239}Pu emits energetic α -particles, care must be exercised in disposing of low-level α -contaminated wastes with low-Z type materials like beryllium ($Z=4$), boron ($Z=5$), and a few other light elements. For example, boron, when exposed to α -particles, can react to release neutrons via the following reaction: $^{10}\text{B} + \alpha\text{-particle} \rightarrow ^{13}\text{N} + \text{n}$. This type of reaction is called an (α, n) reaction. In this situation, nonhazardous, limited range, α -particle radiation can be converted into a more penetrating and hazardous source of neutrons. Because of the low range of α particles, the low Z-material must be in contact with the ^{239}Pu for a significant neutron flux to result.

Table 17-2. A mass-energy balance for Pu-239 decay resulting in the emission of a 5.105-MeV α -particle and a 52-keV gamma-ray.

Mass-Energy Scenario Before ^{239}Pu Decay			
Particle	Rest-Mass or Equivalent Energy ^a (AMU) or (MeV) ^b	Particle Energy Before Decay (MeV)	Total Energy (MeV)
^{239}Pu	239.0521565 AMU 222,675.726 MeV	0.000 MeV ^c	222,675.726 MeV
Total Energy	222,675.726 MeV	0.000 MeV	Before Decay 222,675.726 MeV
Mass-Energy Scenario Following ^{239}Pu Decay			
Released Particle(s)	Rest-Mass or Equivalent Energy ^a (AMU) or (MeV) ^b	Particle Energy After Decay (MeV)	Total Energy (MeV)
^{235}U	235.0439231 AMU 218,942.079 MeV	0.087 MeV ^d	218,942.166 MeV
2 atomic electrons	0.00109716 AMU 1.022 MeV	0.030 MeV ^e	1.052 MeV
α -particle	4.001474918 AMU 3,727.351 MeV	5.105 MeV	3,732.456 MeV
γ -ray photon	0.000 AMU 0.000 MeV	0.052 MeV ^f	0.052 MeV
Total Energy	222,670.452 MeV	Decay energy = 5.274 MeV ^g	After Decay 222,675.726 MeV

a. The energy to mass conversion factor is 931.49432 MeV/AMU.

b. AMU = atomic mass unit = $1.6605402 \times 10^{-27}$ kg; and, MeV = million electron volts (e.g., 1000 keV or 1,000,000 eV).

c. Kinetic energy (KE) of the ^{239}Pu atom is assumed to be zero.

d. Recoil KE of ^{235}U after α -particle emission.

e. Combined KE of two released atomic electrons for this decay scenario. Other values are possible.

f. For this table, the photon energy has been rounded from 51.6 keV to 52 keV. An alternative situation can occur whereby two lower-energy photons can be released instead of a single 52 keV γ -ray (see Table 1).

g. The 5.274 MeV value reported here is based on the calculated mass differences (before and after decay); for example, $222,675.726 \text{ MeV} - 222,670.452 \text{ MeV} = 5.274 \text{ MeV}$. In comparison, Reference 11 reports a decay energy value of only 5.245 MeV for ^{239}Pu .

Table 17-3. Comparison of the average and maximum kinetic energies of atomic electrons (e^-), beta-particles (β), and α -particles released during the decay of several radionuclides (obtained from References 1, 13, and 15).

Radionuclide	Particle	Half-life (years)	Average Energy (keV)	Maximum Energy (keV)
Nickel-59 (^{59}Ni)	e^-	76,000	4 ^a	~8 ^a
Tritium (^3H)	β	12.3	6	19
Plutonium-241 (^{241}Pu)	β	14.4	5	21
Plutonium-239 (^{239}Pu)	e^-	24,100	~15 ^b	— ^c
Iodine-129 (^{129}I)	β	15,700,000	40	150
Carbon-14 (^{14}C)	β	5,715	49	156
Technetium-99 (^{99}Tc)	β	213,000	85	293
Iodine-131 (^{131}I)	β	0.022	180	806 ^d
Cesium-137 (^{137}Cs)	β	30.1	195	1,176
Potassium-40 (^{40}K)	β	1,270,000,000	541	1,330
Phosphorous-32 (^{32}P)	β	14.3	694	1,710
Gadolinium-148 (^{148}Gd)	α	75	3,180	3,180
Uranium-238 (^{238}U)	α	4,470,000,000	4,190	4,200
Radium-226 (^{226}Ra)	α	1,600	4,770	4,780
Neptunium-237 (^{237}Np)	α	2,140,000	4,770	4,790
Thorium-230 (^{230}Th)	α	80,000	4,660	4,690
Plutonium-241 (^{241}Pu)	α	14.4	4,890	4,900
Actinium-227 (^{227}Ac)	α	22	4,940	4,950
Protactinium-231 (^{231}Pa)	α	32,800	4,960	5,060
Plutonium-239 (^{239}Pu)	α	24,100	5,150	5,160
Thorium-228 (^{228}Th)	α	1.9	5,390	5,420
Americium-241 (^{241}Am)	α	433	5,320	5,490
Plutonium-238 (^{238}Pu)	α	87.7	5,490	5,500
Californium-252 (^{252}Cf)	α	2.65	5,930	6,120
Curium-242 (^{242}Cm)	α	163	6,100	6,120
Einsteinium-252 (^{252}Es)	α	1.3	6,300	6,640

a. The data for ^{59}Ni represents Auger or atomic electrons and not electrons emitted from the nucleus (i.e., beta particles).

b. Estimated from mass-energy balance equations.

c. A few very energetic electrons (~1 MeV) can be released from ^{239}Pu decay, but these events are extremely rare.

d. Beta particle intensity for ^{131}I is 90.4% at 606 keV and 0.6% at 806 keV.

Table 17-4. Comparison of the radiotoxicity of several important radionuclides including ^{239}Pu .
Obtained from Appendix 2 of Reference 7.

Radiotoxicity		Species
Very high	Group 1	^{241}Pu , ^{242}Cm , ^{241}Am , ^{237}Np , ^{238}Pu , ^{239}Pu , ^{240}Pu , ^{242}Pu
High	Group 2	^{60}Co , ^{90}Sr , ^{94}Nb
Moderate	Group 3	^{14}C , ^{63}Ni , ^{137}Cs
Low	Group 4	^3H , ^{59}Ni , $^{99\text{m}}\text{Tc}$, ^{99}Tc , ^{129}I

CHEMICAL AND PHYSICAL CHARACTERISTICS

Plutonium (element 94) is a member of the actinide series of elements (i.e., elements 89 through 103) of the periodic table. Its chemical characteristics are similar to that of the rare earth elements (a series of metallic elements ranging from lanthanum — element 57 — through lutetium — element 71). In particular, plutonium is in the same chemical family as the rare earth element samarium. Plutonium is similar to uranium, neptunium and americium in that all of these elements have four possible oxidation (valence) states (i.e., +3, +4, +5, and +6). In aqueous solutions, plutonium may exist in all four valence states. The +4 state is the most stable oxidation state for plutonium. Plutonium occurs in six different crystalline structures, or allotropic states, each existing within a unique temperature range. The existence of these six allotropes complicates its metallurgical properties. Also, the metallurgical properties of plutonium are very sensitive to the presence of impurities. Table 5 summarizes some of the most important chemical properties of plutonium. In the absence of surface oxidation, plutonium is a silvery-white metal with the appearance of nickel.¹¹ As the metal is oxidized (by air or water), plutonium acquires a bronze-like color, followed by a gunmetal-blue color, and finally a dull black or green at complete oxidation. Besides oxygen, plutonium metal is chemically attacked by ammonia (NH₃), nitrogen (N), hydrogen (H), halogens (e.g., fluorine, chlorine, bromine, iodine), carbon monoxide (CO), and carbon dioxide (CO₂). Plutonium reacts with many elements to form at least 90 known binary compounds.¹⁸ Some of the most well known plutonium compounds are: plutonium oxides (PuO, Pu₂O₃, PuO₂, etc.), plutonium hydrides (PuH₂, PuH₃, etc.), plutonium fluorides (PuF₃, and PuF₄ both highly insoluble in water and acids), plutonium chlorides (PuCl₃ and PuCl₄), plutonium carbides (PuC, Pu₂C₃, and PuC₂), plutonium nitrides (PuN, Pu₂N₃, and PuN₂), plutonium sulfides (PuS, Pu₂S₃, etc.), and many others. Most of these compounds are important in the production of plutonium-based nuclear fuels (e.g., mixtures of PuO₂ and UO₂), uranium and plutonium separation processes, or the production of plutonium metal. Metallic plutonium appears to be more chemically reactive than metallic uranium at similar temperatures.² Because of the highly electropositive nature of plutonium, the metal is soluble or attacked by many acids (e.g., HCl, HBr, 72% HClO₄, 85% H₃PO₄, trichloroacetic acid, and other acids).

Plutonium is a highly reactive metal that combines readily with oxygen to form plutonium oxide (PuO₂). Plutonium is also hygroscopic in that it combines with water or water vapor; however, it reacts only slowly with water at room temperature and slightly faster at the boiling point.¹¹ Plutonium metal, in the form of a powder or finely divided material, such as machine filings, is pyrophoric at temperatures of about 753 K (480°C).⁴ Fires involving plutonium produce plutonium oxide smoke particles. Therefore, machining and manufacturing activities of plutonium are usually conducted in a dry inert gas atmosphere of helium or argon and sometimes in a vacuum.

Both ²³⁹Pu and ²⁴¹Pu can be fissioned with slow neutrons, and are called fissile isotopes. These two isotopes can form critical assemblies (a mass of material that can sustain a fission chain reaction) under certain conditions of geometry and mass. Because plutonium is fissionable, careful planning must be exercised in its handling and machining. The critical mass of plutonium varies over a wide range, depending on such factors as geometry, reflectors, material mass or density, and the effect of diluents. For example, in an optimal configuration the minimum critical mass for ²⁴¹Pu is 260 g in a water solution, compared with ²³⁹Pu with a minimum critical mass of 511 g (fully reflected geometry), or 10.2 kg for a bare metal assembly.¹⁹ In fact, plutonium in a liquid solution is more likely to become critical than solid metallic plutonium.

Table 17-5. Physical properties of plutonium.

Physical property (units)	Plutonium data	Temperature Range (K)
Melting point (K) ^a	913 ^b	
Boiling point (K) ^a	~3500	
Heat of fusion at melting point (kJ/mole)	2.84 ^c	
Heat of vaporization (Kcal/mole)	80.49 ^d	
Solid density (g/cm ³)	19.84 to 15.92 ^e	
Liquid density (g/cm ³)	— ^f	
Oxidation potential for the first electron (V)	6.06 ^c	
Ionization potential (eV)	— ^f	
Work function (eV)	— ^f	
Crystal forms and temperature ranges ^e	alpha (monoclinic)	< 395 K
	beta (monoclinic)	395-479 K
	gamma (orthorhombic)	479-592 K
	delta (cubic)	592-724 K
	delta prime (tetrahedral)	724-758 K
	epsilon (cubic)	758-912 K

a. Note that 0 K equals -273.15°C or -459.67°F.

b. The melting point of plutonium varies with its initial crystalline phase designation and lattice structure are listed as follows: alpha (α) monoclinic 914 K, Beta (β) monoclinic 395–480 K, gamma (γ) orthorhombic 480-592 K, delta (δ) cubic 592-724 K, delta prime (δ') tetrahedral 724–749 K, epsilon (ε) cubic 749–913 K. Data obtained from References 20 and 2.

c. Obtained from the Sargent-Welch Table of Periodic Properties of the Elements.

d. Obtained from Reference 18.

e. The density of plutonium varies with crystalline form: α monoclinic 19.84 g/cm³; β monoclinic 17.70 g/cm³; γ orthorhombic 17.14 g/cm³; δ cubic 15.92 g/cm³; δ' tetrahedral 16.00 g/cm³; and ε cubic 16.51 g/cm³.

f. Information not available.

PLUTONIUM-239 PRODUCTION IN NUCLEAR REACTORS

Plutonium-239 is an artificially produced radionuclide that does not normally exist in nature. It is created in nuclear reactors by a single neutron capture of ^{238}U (e.g., $^{238}\text{U} + n \rightarrow ^{239}\text{U} \rightarrow ^{239}\text{Np} + \beta + \gamma \text{-ray} \rightarrow ^{239}\text{Pu} + \beta + \gamma \text{-ray}$). The majority of the plutonium that is produced in nuclear reactors is contained within the spent fuel elements. The plutonium isotopes most commonly found in nuclear reactors consist of isotopes ranging from ^{236}Pu to ^{244}Pu ; but mainly isotopes ^{239}Pu , ^{240}Pu , ^{241}Pu , and ^{242}Pu , with ^{239}Pu representing the most abundantly produced plutonium isotope (on a mass basis).

Due to the high neutron absorption and fission cross-sections of ^{239}Pu (radiative capture cross-section: $\sigma_{\gamma}=274 \text{ barns} = 274 \times 10^{-24} \text{ cm}^2$, and the average fission cross-section of $\sigma_f=698 \text{ barns}$ at 0.0253 eV), a significant fraction of the ^{239}Pu that is produced in a reactor is also destroyed. The amount of ^{239}Pu existing in a reactor will depend upon a complex function of the initial fuel composition, reactor power, fuel burnup, and the fuel irradiation history. Table 6 shows an ORIGEN2²¹ calculated amount of ^{239}Pu in a typical 3,200-MWt light-water reactor (LWR) initially fueled with 100 Mt of ^{235}U (3.3%) + ^{238}U (96.7%), and operated for 3 years.

The primary source of plutonium is through reprocessing irradiated nuclear fuel; however, a sizable amount can now be recovered from decommissioned weapons. Figure 1 shows a comparison of the fission product yields from ^{239}Pu and ^{235}U due to thermal-neutron-induced fission as a function of mass. A significant difference exists in these two yield curves, with ^{239}Pu producing more heavier fission products than fission of ^{235}U for isotopes of mass greater than about 150. This region includes the rare earth elements, which represent some important reactor poisons due to their large thermal neutron cross-sections.¹¹

Table 17-6. Plutonium-239, total plutonium, total actinide, and total fission product inventories calculated as a function of decay time following a 3-year irradiation in a 3,200-MW thermal power LWR producing a total fuel burnup of 33,400 MWtD/MTU. Results are presented in terms of millions of curies (first list) and kilograms (second list). These results are based on an ORIGEN2 calculation for a 100 MTU (3,300 kg ^{235}U + 96,700 kg ^{238}U) pressurized water reactor.

Radioactive Material	Reactor Core Inventory (millions of curies)					
	Completion of Fuel Irradiation T_0	Decay Time (years) Following Reactor Irradiation				
		T_0+1	T_0+5	T_0+10	T_0+50	T_0+100
^{239}Pu	0.0288 ^a	0.0293 ^a	0.0293	0.0293	0.0293	0.0293
All Pu Isotopes	58.8	15.7	13.0	10.3	1.73	0.340
All Actinides	3,900.0	17.6	13.4	10.7	2.25	0.836
All fission products (FPs)	15,000.0	200.0	46.4	31.6	11.3	3.51
Actinides plus fission products	18,900.0	217.6	59.8	42.3	13.6	4.34
Radioactive Material	Reactor Core Inventory (kilograms)					
	Completion of Fuel Irradiation T_0	Decay Time (years) Following Reactor Irradiation				
		T_0+1	T_0+5	T_0+10	T_0+50	T_0+100
^{239}Pu	464.0 ^a	472.0 ^a	472.0	471.0	471.0	471.0
All Pu Isotopes	857.0	859.6	833.8	807.2	720.8	703.0
All Actinides	96,380.0	96,380.0	96,380.0	96,380.0	96,380.0	96,380.0
All fission products (FPs)	3,620.0	3,620.0	3,620.0	3,620.0	3,620.0	3,620.0
Actinides plus fission products	100,000.0	100,000.0	100,000.0	100,000.0	100,000.0	100,000.0

a. The inventory of ^{239}Pu goes up after reactor shutdown due to the decay of precursors ^{239}U and ^{239}Np .

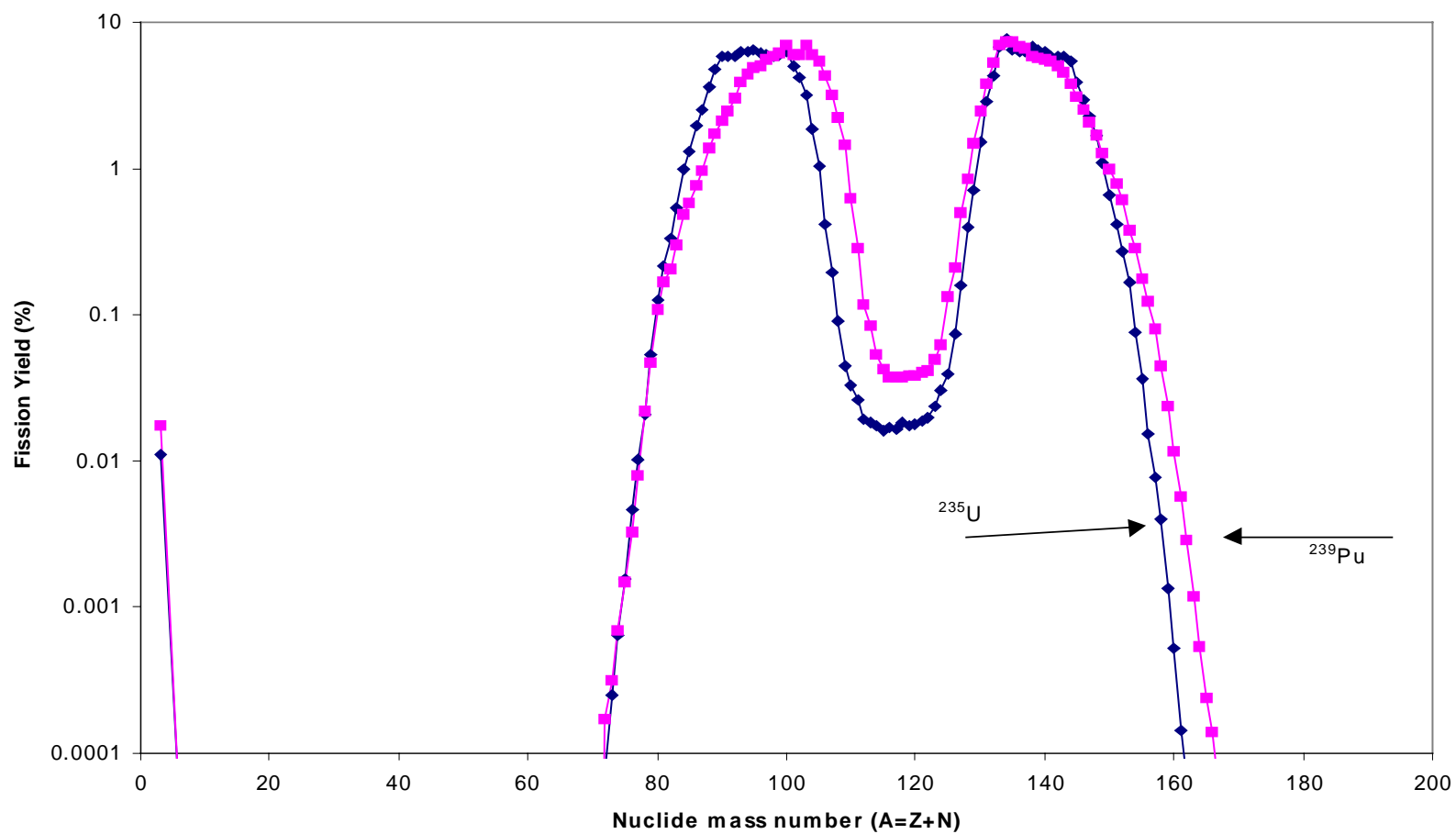


Figure 17-1. Fission product yield curve for thermal fission of ^{235}U and ^{239}Pu . The curve with diamonds represents ^{235}U , and the curve labeled with boxes represents ^{239}Pu .

WASTE AND DISPOSAL DATA ON PLUTONIUM-239

Nuclear Reactors

In a typical nuclear fuel cycle, plutonium normally exists as an oxide material (e.g., PuO_2), with the plutonium isotopic abundance consisting of ~44–70% ^{239}Pu and ~20–30% ^{240}Pu ; with various amounts of uranium isotopes (depending on the particular fuel cycle). For example, mixed oxide (MOX) fuels consist of various blends of uranium, plutonium, and/or thorium; however, visually there is no difference between a UO_2 fuel pellet and a MOX fuel pellet containing a mixture of UO_2 , PuO_2 , and ThO_2 . Even in a 3,200-MW thermal light-water reactor (LWR) initially fueled with only ^{235}U and ^{238}U , there will eventually be produced some 860 kg of plutonium and 470 kg of ^{239}Pu near the end its core life-time (see Table 6). Larger quantities of plutonium can exist in Liquid Metal Fast Breeder Reactors.

Recycling of the ^{239}Pu present in expended LWR fuel elements can only be accomplished through reprocessing of this fuel; that is, dissolving the fuel in acids and chemically separating the plutonium from the other heavy metals. Presently this is not being performed on LWR fuel rods, but is accomplished on some naval fuels. Instead, all irradiated LWR fuel rods are ultimately destined for permanent geologic storage at a DOE repository for spent radioactive fuel; possibly the Yucca Mountain site.

Almost all of the ^{239}Pu that is produced in reactors remains in the fuel elements and does not end up in a LLW site. However, there is a very small fraction of the total reactor inventory of this nuclide that does end up as low-level waste, primarily associated with contaminated hardware, clothing, etc., as discussed in the next section.

Medical, Academic Institutions, and Industrial Plutonium-239 Wastes

Pure sources of ^{239}Pu have only been available since about 1960.¹¹ As mentioned before, ^{239}Pu is available from Oak Ridge National Laboratory in the following units: (a) >99% ^{239}Pu at \$1.25 per mg, and (b) 99.99% pure ^{239}Pu at \$4.80 per mg. A packaging fee of \$2,000 to \$3,000 and freight charges are extra.

Plutonium has a very long radiological half-life and, as stated in an earlier section, emits a high-energy α particle. It also has a long biological half-life (the length of time for an organism to rid itself of one-half of a given dose of the nuclide through biological means, such as excretion or urination). Thus, there are no significant uses for this nuclide in the treatment of disease.

There have been several animal-based studies of the effects of ^{239}Pu in the body. These have been conducted in an effort both to measure the metabolism and distribution of this nuclide within the human body and to measure the effect of the nuclide on the human body. These medical studies have resulted in ^{239}Pu -laden waste products.

Additionally, studies have been and are being conducted to measure the nuclear properties of ^{239}Pu . These, too, have resulted in the production of ^{239}Pu -laden waste products. However, the principal uses for this nuclide are as the fissile agent in nuclear weapons and as fuel for nuclear reactors.

Disposal Data on Plutonium-239

^{239}Pu has been deposited in three LLW sites in Richland, Washington; Barnwell, South Carolina; and Beatty, Nevada over a period of several years. (Note: this nuclide has also been disposed of by Envirocare in their LLW site in Utah, but only since January 1998 – approximately 0.03 Ci total activity has been deposited. Data from this LLW site are not yet in the database and, therefore, are not included in the discussion in this section.) Data concerning the activity, sources, and waste types for this nuclide have been obtained from the Manifest Information Management System (MIMS) Database.^b Figure 2 shows the total annual ^{239}Pu activity deposited in the three sites for each year between 1986 and 1998 (Note: the data for 1998 were not complete at the time of publication). In only 3 years did the deposition of ^{239}Pu exceed one Ci, 1986, 1992, and 1993. The general trend has been for less and less of this nuclide to be deposited in the LLW sites.

Figure 3 shows the activity of ^{239}Pu deposited in these three sites as a function of waste source. Approximately one-half of the total activity of ^{239}Pu deposited in LLW sites was from utility sources. The other approximate one-half was from industry and government sources, with only minor contributions from academic and medical sources.

Figure 4 shows the activity of ^{239}Pu deposited in the Richland and Beatty sites as a function of waste type. The waste disposal manifest for the Barnwell site did not break down the waste by type, which is unfortunate since more than 50% of the total activity of ^{239}Pu is deposited in this site. An additional 22% of the total deposited activity was likewise not broken down by waste type for the other two sites. Thus, more than 70% of the total ^{239}Pu deposited activity was not listed by waste type. None of this activity was included in the data shown in Figure 4, which, therefore, only illustrates the waste types for less than 30% of the total deposited activity in the LLW sites.

As shown in this figure, more than half of the activity is in the form of dry solids. In addition, approximately one-fourth of the total activity is in the form of dry active waste, which is a combination of compacted and noncompact active waste. Dewatered resins and noncartridge filter media add another 10% and the remainder is scattered among the other four waste types.

The data in Figures 3–5 are only for years since 1986. This is because data before 1986 are not available in the MIMS database. However, ^{239}Pu depositions have taken place since as early as 1962. ORNL has published an annual report that documents historical LLW inventories, including depositions starting in 1962.²² The data for individual nuclides are not available in the reference, but some information can be extracted from the data that are presented. Table 4.17, in the reference, contains total deposition data for each of the three LLW sites. These data are of the total LLW deposition volume for each year, starting in 1962. Although this table does not break the yearly depositions down according to radionuclide, Table A.3 (in Reference 22) contains historical conversion factors (from volume to activity) for each radionuclide, including ^{239}Pu . These conversion factors appear to be only suitable for trend indications. A comparison between the derived ^{239}Pu depositions (from Reference 22) and the data from the MIMS database, for 1986–1995, indicate that the MIMS values generally are higher, by as much as a factor of 10. However, the Reference 22 data are useful for evaluating deposition trends. Total (including ^{239}Pu) depositions peaked in 1980, and then generally decreased. By 1996, the total deposition was approximately one-tenth that for the peak year. Possible reasons for this decrease include the decline in the construction of commercial nuclear power plants and the decline in the production of weapons materials, which resulted from the strategic arms limitation treaties.

b. Information provided through the DOE waste management system maintained by the National Low-Level Waste Management Program, Idaho National Engineering and Environmental Laboratory (INEEL).

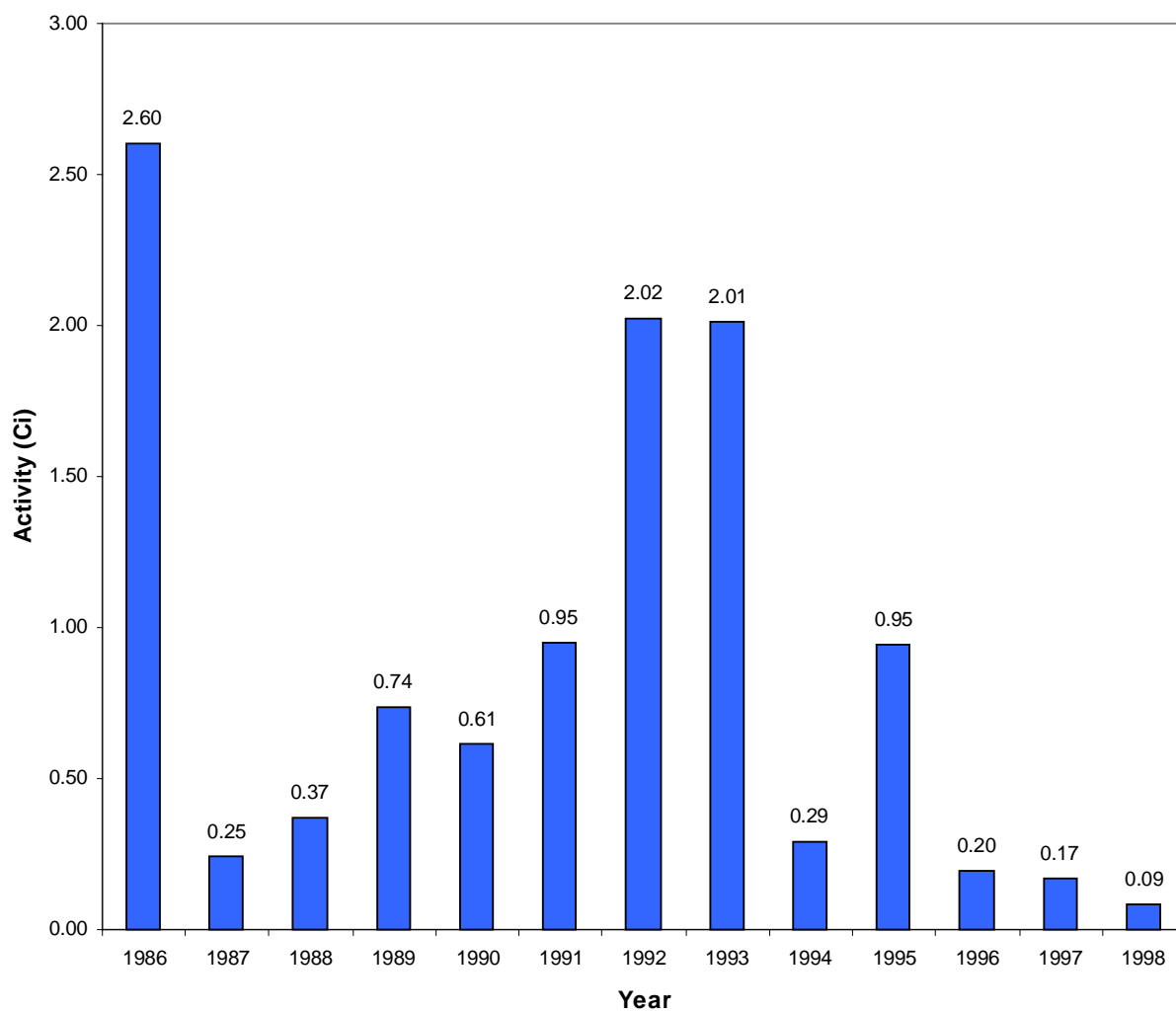


Figure 17-2. Pu-239 activity deposited at the Richland, Beatty, and Barnwell sites by year.

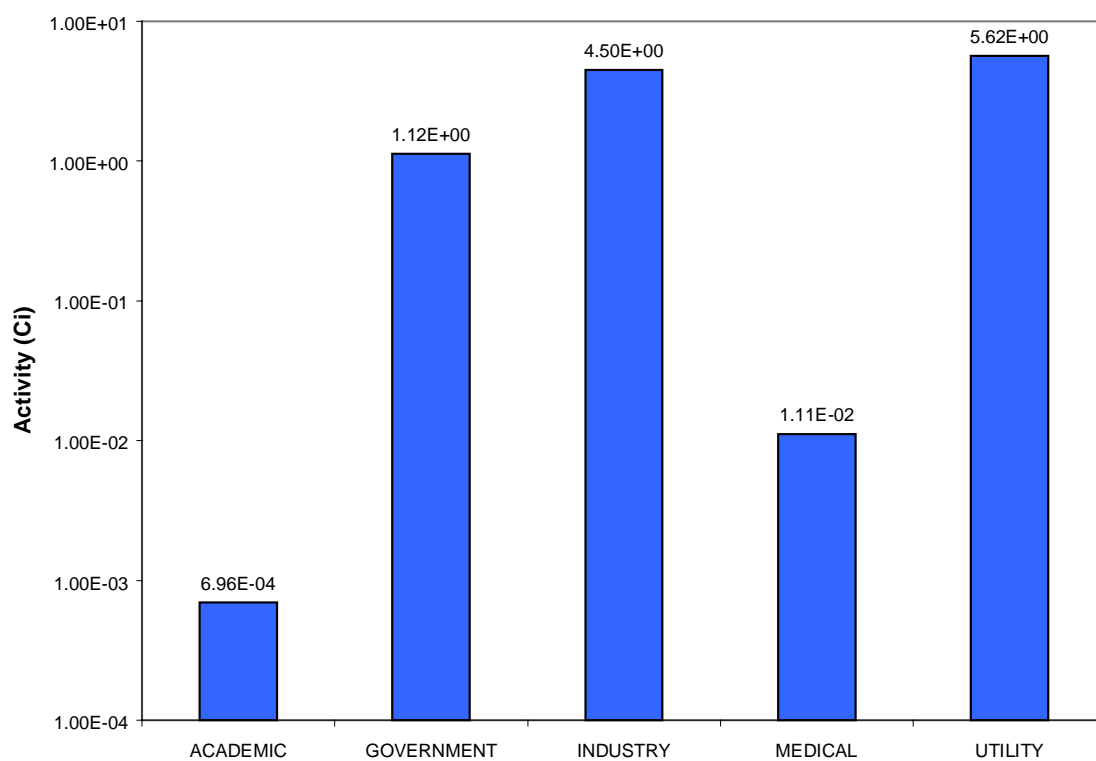


Figure 17-3. Pu-239 activity deposited at the Richland, Beatty, and Barnwell sites by source.

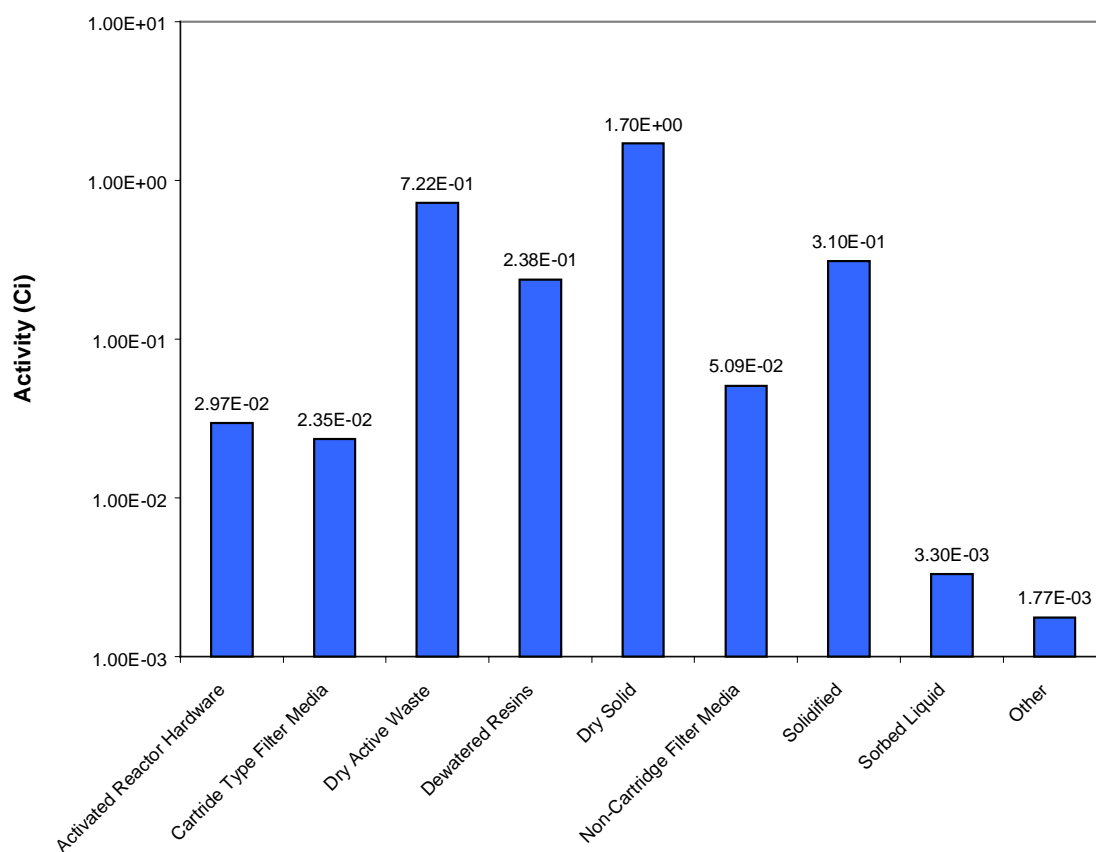


Figure 17-4. Pu-239 activity deposited at the Richland and Beatty sites by waste type.

BEHAVIOR OF PLUTONIUM IN THE ENVIRONMENT

Plutonium in Soils

Understanding the behavior of plutonium in the soil is important in predicting the dissolution of radioactive waste in low-level waste disposition (e.g., burial) and the possible transport of plutonium through the biosphere. It is important to understand the interactions between radionuclides and various media along the path to the biosphere, whether disposal is in deep or shallow rock caverns or in shallow overburden facilities.

Plutonium does not naturally occur in soils; however, it is present in many soils because of a variety of reasons. The primary sources of environmental plutonium include (a) releases from research facilities, (b) nuclear weapons testing, (c) waste disposal, (d) nuclear weapons production facilities, (e) experimental operations, and (f) accidents. Nevertheless, the major source of plutonium in most soils is primarily due to radioactive fallout from weapons testing.²³

Soil concentrations of ²³⁹Pu have been measured at numerous locations around the world. The results from three of these studies are summarized here. The concentration of this radionuclide at various locations on the Hanford DOE Site (Pacific Northwest National Laboratory) ranged from 0.01 to 0.09 pCi/g.²⁴ Radionuclide contamination of soils in Korea were measured by scientists from the Korea Institute of Nuclear Safety.²⁵ The results from this Korean study indicate that of the radionuclides included in the study, the Pu isotopes had lower (activity) concentrations and the concentrations of ²³⁹⁺²⁴⁰Pu were much greater than those of ²³⁸Pu. The ²³⁹⁺²⁴⁰Pu concentrations ranged from 0.18 to 1.85 Bq/kg (4.9–5 x 10⁻³ pCi/g). These concentrations tended to be higher for soils containing more organic matter or clay content, as well as soils from regions with higher mean annual precipitation.

Past investigations have indicated that plutonium sources from fallout and industrial releases behave in a similar fashion, and that when soluble or insoluble forms of organic matter are present in either alkaline or acid soils, any contaminating plutonium in the soil has the potential of high mobility.²⁴ In addition, the important factors affecting plutonium chemistry and, therefore, mobility and sorption in soil, are (a) soil pH, (b) clay content, (c) calcium carbonate content, and (d) organic matter content.²⁴ For example, studies of plutonium concentrations due to fallout on soils from Ohio (a humid area with high glacial lime tills) showed a significant correlation with clay content. However, experimental investigations showed that ~95% of the plutonium in solution at a pH of 6.5 could be removed by calcium-saturated clays.

An additional factor affecting plutonium chemistry and binding capabilities to soil is microbial activity. It has been suggested that up to 10% of the plutonium present in soil may become bound to or associated with organic compounds present in the soil.²⁴ Even plutonium added to a culture medium as PuO₂ (as very small particles) can be assimilated by common soil fungi. The diffusion of plutonium in soil is also affected by the sorption characteristics of the various chemical forms present. However, plutonium may be more mobile in soil than diffusion theory can explain. In particular, micro-organism activity can increase plutonium mobility in soil. Microbiological and plant actions can influence plutonium mobility in soil, and these factors will be of increased importance as larger quantities of plutonium become bound to living and dead organic materials. It has also been noted that the movement of PuO₂ was approximately 100 times faster than that of Pu(NO₃)₄.²⁴ Also, physical processes (e.g., wind and water transport) may be more important than chemical processes in transporting plutonium to vegetation.

It appears that in the majority of soil types, and especially those that contain significant quantities of clay, soluble forms of plutonium can be adsorbed rapidly by the soil. Several studies have been made

on the vertical distribution of plutonium in soil profiles contaminated as a result of nuclear fallout and accidental releases.²⁴ Generally, these studies show slight concentrations in surface soils relative to subsurface soils. In fact, it is reported in Reference 24 that more than 95% of the plutonium present was in the top 5-cm layer. Several authors report a decreasing exponential relationship between plutonium concentration and soil depth. In the case of the Trinity nuclear test site in New Mexico, consisting of sandy loam soil, about 50% of the plutonium residing in the top 0 to 5-cm layer of soil has moved downward to the 5 to 20-cm layer over a 20-year period.²⁴ It has also been noted that the mobility of plutonium in soil profiles can be expected to be the greatest when the chemical form of plutonium is largely as the oxide (PuO_2), or when the soil is low in clay content, high in soluble organic material, and subject to vegetation cover. In farming areas, the effect of plowing on plutonium movement has to be considered.

The soil retention parameter represents the solid/liquid partition coefficient and is denoted by the symbol K_d (i.e., a distribution coefficient). K_d is defined by $K_d = C_s/C_l$, where C_s is the plutonium concentration in the soil (solid) and C_l is the plutonium concentration in the groundwater (liquid). This empirical model combines the soil retention mechanisms into a simple linear partition relation between the soil and the surrounding groundwater. This model assumes that the C_s and C_l are in equilibrium with each other; that is, they are not changing with time. Using this definition for K_d , it follows that the larger the value (that is, the higher the radionuclide concentration in the soil relative to that in the groundwater) the slower the migration of the radionuclide relative to groundwater flow. Therefore, K_d can be thought of as a measure of the amount of “holdup” in the soil.

Typically, K_d is measured under laboratory conditions, using samples that are relatively homogenous, and where it can be ensured that equilibrium conditions are met. Applying these values to specific soils in the field can be difficult because actual soils are generally inhomogeneous and there are uncertainties as to how long it takes for the plutonium concentrations in the soil and groundwater (C_s and C_l) to be in equilibrium with each other. Therefore, one must be careful to ensure that soil samples used in the laboratory studies are as closely representative of the field as possible. However, even though the soil samples do not precisely match conditions in the field, K_d values from these laboratory studies can be used in computer models that extrapolate data from laboratory experiments and field studies. In addition, some experiments are conducted using intact field samples to validate the laboratory K_d values and to study the effects due to soil nonhomogeneity.

Scientists at the Laboratory of Nuclear Medicine and Radiation Biology of the University of California at Los Angeles measured the K_d for several radionuclides, including ^{239}Pu .²⁶ On a relative basis, they determined that the K_d values were in the following order: $^{237}\text{Np} < ^{239}\text{Pu} < ^{241}\text{Am}$. Thus, ^{239}Pu is transported more easily than ^{241}Am but is more easily retained in the soil than ^{237}Np , on a relative basis. These coefficients were measured over a wide variety of soils and K_d for ^{239}Pu ranged from less than 50 to nearly 7,000. An earlier investigation of K_d was performed by scientists associated with the Pacific Northwest Laboratory.²⁷ In this study, the distribution coefficient for plutonium(IV) was measured to be of the order of 10,000. A more recent study was performed by scientists at the Japan Atomic Energy Research Institute in Tokai, Japan.²⁸ The distribution coefficient for plutonium(IV) was measured in the laboratory for a variety of sandy soils to be of the order of 10,000 mL/g. However, an *in-situ* measurement of K_d was made, which resulted in a much smaller value - of the order of 1,000 mL/g. The difference between the laboratory and the *in-situ* values of K_d are believed to be due to differences in the oxidation state of the plutonium. This illustrates the care that must be taken in using laboratory values for this parameter in predicting movement of plutonium in the field.

These large values of K_d for ^{239}Pu and ^{241}Am are consistent with the results of a study conducted by scientists associated with the Institute for Radiation Protection in Neuherberg, Germany.²⁹ This study involved the vertical migration into soil of several radionuclides resulting from fallout associated with the

nuclear reactor accident at Chernobyl, Ukraine. The results of their study indicated that the vertical migration of these two radionuclides (^{239}Pu and ^{241}Am) were of the order of <1 cm/year, which was much less than the other radionuclides included in the study. A similar study, conducted by scientists associated with the Academy of Sciences of Belarus, produced comparable results.³⁰ In this later study, it was also concluded that “the tendency for mobility of radionuclides (is) to decrease with increasing depth.” In the Pacific Northwest National Laboratory study documented by Reference 24, it was determined that ^{239}Pu has a tendency to “strongly sorb to soil and . . . (be) readily retained in surface soil.” Additionally, the presence of soil microbes can enhance the migration of Pu in soil, though this effect appears to be small in the soil samples from the Hanford Site.

Another aspect of the behavior of ^{239}Pu in soils is related to mechanisms of removal (redemption). Several studies have been made of soil remediation, of which the following three are typical examples. Nishita and Hamilton measured the effect of organic material and soil pH on the extractability of ^{239}Pu .³¹ The results from this study indicate that removal of organic material from the soil appreciably increased the amount of ^{239}Pu that could be extracted from a kaolinitic soil under acidic pH conditions. The presence or absence of organic material did not have an appreciable effect under alkaline pH conditions. A study was conducted by scientists at the Los Alamos National Laboratory into the ability of bioreductive processing to remove actinide contaminated soils.³² For this study, soils from several DOE sites were collected and a proprietary process, developed by MBX Systems, Inc. was used to extract the actinides, including Pu. The results from this study indicated that up to 97% of the Pu could be removed from these contaminated soils.

Although under normal soil conditions, plutonium tends to be fixed in place and not migrate significantly, there are conditions under which this can change. In one of these conditions, substantial amounts of colloidal material are suspended in the ground water. This is discussed in more detail in another paragraph. In addition, the migration of plutonium can also be significantly affected by the presence of chelates, which form complexes with the plutonium compounds that can be significantly more soluble than the plutonium compound alone. Several examples are included in this paragraph. A detailed discussion of the effects of chelates on the solubility of plutonium is too complex to be discussed in this report. The interested reader is referred to the literature for more specific information. Scientists at the University of California at Los Angeles conducted an experimental study on the effects of fulvic and humic acids on the extractability of plutonium.³³ The extractability of plutonium (which is a measure of the mobility of plutonium in soil) was shown to be strongly influenced by the soil pH, the relative amounts of organic and inorganic phases in the soil, and on the concentration of the complexing agent(s). The authors concluded that under normal soil conditions, plutonium is relatively immobile. However, under certain conditions, the extractability of plutonium can be enhanced significantly due to the formation of complexes with fulvic and humic acids.

Scientists from Pacific Northwest National Laboratory have made in-situ measurements of the migration of various radionuclides, including ^{239}Pu , from the shallow waste burial site at Maxey Flats, Kentucky.³⁴ The authors conclude that the presence of organic complexing agents, especially ethylene diamine tetraacetic acid (EDTA), can enhance the mobility of plutonium. The measurements supporting this conclusion are somewhat masked by the fact that this site is in a region of high rainfall and some of the transport may have been due to effects other than groundwater transport. As with the Reference 33 study, the mobility of plutonium is complex and is strongly influenced by such factors as soil pH and soil organic content.

Plutonium in Water

Plutonium contamination in the environment began in 1945 with the nuclear weapons test at Alamogordo, New Mexico and the nuclear attack at Nagasaki in Japan. The climatic conditions of Alamogordo are very dry, but Nagasaki is very wet, with annual precipitation typically ranging from 60 to 110 inches. While a large portion of the fallout plutonium was retained in the upper soil level, even after 40 years, some did migrate to the water table. This was confirmed by the detection of trace amounts of plutonium in seeping groundwater at Nagasaki.²⁸ It was shown that the mobility of plutonium(IV) in groundwater is increased by the quantity of dissolved organic carbon in the water, which is generally known to have a high potential to chemically associate with plutonium.

Plutonium concentrations and oxidation states were measured in water collected from the shallow aquifer in the Mortandad Canyon near the Los Alamos National Laboratory.³⁵ The source of the plutonium in the aquifer was treated waste that had been released in the canyon for approximately 20 years. Concentrations of ^{239,240}Pu in the water were measured to be in the range of 0.018 to 5.4 pCi/L, with the higher concentrations associated with samples taken near the discharge site. It was judged that the fine particulate matter in the water is a poor adsorber of plutonium and that there was a complexing agent in the water that helped maintain the plutonium in solution and mitigated against it being adsorbed onto the adjacent soil. There was almost no correlation between plutonium concentrations in the water and the presence of dissolved organic material.

Low levels of radioactivity were in the water routinely discharged from the N-Reactor at Hanford into a rock-lined crib and overflow seepage trench located near the reactor site and on a bluff overlooking the Columbia River.³⁶ While most of the radioactivity was retained, by design, in the crib and seepage trench, some of the nuclides, including some plutonium species, did migrate to the river. The concentrations of ^{239,240}Pu in springs that feed the river ranged from 0.008 to 0.030 pCi/L (compared with concentrations in the trench water ranging from 24 to 58 pCi/L, or more than 3 orders of magnitude higher). Of the plutonium that reached the spring water, ~90% of it was in oxidation states V and VI, which tend to be more mobile oxidation states for plutonium. A follow-on study was conducted to investigate the oxidation states of plutonium in groundwater.³⁷ This study concluded that the plutonium enters the trench in a high oxidation state (V, VI) and then is rapidly reduced to lower oxidation states (III, IV) and adsorbed onto sediments in the trench. The fraction of plutonium that remains in the higher oxidation states enters the groundwater and is further reduced, though not as rapidly as in the trench, and partially removed by adsorption onto soil.

As part of the characterization studies for siting a high-level radioactive waste repository at Yucca Mountain, the solubility of plutonium and other actinides were measured in representative groundwater samples from the region.³⁸ The dependence of solubility on temperature (25–90°C) and pH (5.9–8.5) was measured. Plutonium solubility increased with increasing temperature. The principal oxidation states for the dissolved species were V and VI, and as the pH increased, Pu(V) increased and Pu(VI) decreased, though the overall plutonium concentration did not vary significantly.

Even though plutonium is known to be relatively immobile in groundwater as a result of its being adsorbed onto soil particles, this nuclide could become associated with colloids, thus becoming more mobile. Colloids are small (generally submicrometer size) particles that are often found suspended in groundwater. Under some conditions, plutonium can become adsorbed onto these colloidal particles, enhancing its mobility in groundwater.³⁹ In the Reference 39 study, plutonium has been measured in test wells located 1.3 km from the source, an underground nuclear weapons test site located at the Nevada Test Site. Colloidal transport, which is suspected to have contributed significantly to this transport, adds another dimension to the traditional view of radionuclide transport, which usually is considered to be dominated by a stationary phase associated with aquifer solids and a mobile phase associated with

dissolution of the nuclide in the water. Because colloid groundwater concentrations are typically quite low, the contaminants most likely to be transported by colloidal materials are of extremely low solubility and strongly partition to mobile nonaqueous phase materials.⁴⁰ The understanding of colloidal transport is relatively immature and Reference 40 recommends that additional experimental and modeling studies be conducted to enhance the understanding of this phenomenon.

Plutonium in Plants

One of the principal pathways for exposure of humans to ²³⁹Pu is for the plutonium to contaminate vegetation. Then, either animals can ingest the contaminated plants, becoming contaminated themselves, and humans eat the animals, or humans can directly ingest the contaminated plants. Either way, humans become contaminated with this radionuclide.

Plant contamination can occur from either of two phenomena. First, the plants grow in contaminated soil and take up the plutonium via the roots. Second, the plutonium becomes airborne and is deposited directly onto the plant surfaces, where it can be absorbed into the plant structure or remain on the surface. The airborne pathway is the subject of a later section. The first is the subject of this section.

Concentration ratios are used to describe the tendency of plants to extract radionuclides from the soil and deposit them in the plant structures. The vegetation-soil concentration ratio, C_R is defined as the concentration of (for example) ²³⁹Pu in the plant mass (Ci/g) divided by the concentration of the radionuclide in the soil in which the plant was grown (Ci/g). A number of factors have been identified as being of importance with plutonium absorption and the soil-plant relations. These factors include (a) the vegetation type, (b) species, (c) age and status of the plant, (d) soil pH, (e) positive ion-exchange capacity, (f) organic matter and/or clay content, (g) plutonium chemical form added to the soil, and (h) the duration of the contamination. Some authors have remarked that results obtained under experimental conditions differ substantially from field results, mainly due to plutonium uptake from the soil. It is likely that the chemical and physical form of the plutonium source material does affect the plant concentrations derived from the atmosphere, and that the plutonium derived from the soil is controlled by the soil chemistry. Most researchers have concluded that root uptake of plutonium is limited, especially when contrasted with atmospheric sources of plutonium.²³

In 1991, the trees on the dikes surrounding the Savannah River Laboratory seepage basins were sampled and analyzed to measure the concentration ratio of various radionuclides, including ^{239,240}Pu.⁴¹ Of the radionuclides included in this study, plutonium tends to have a smaller concentration ratio, indicating that uptake via the roots is relatively small. The concentration ratios for ^{239,240}Pu ranged from 0.00012 to 0.0073. In relative terms, these nuclides tended to concentrate more readily in hardwood trees than in pines and the distribution between leaves and the wood parts was relatively equal.

A literature study was made of the uptake of various food plants grown in soils contaminated with various radioactive actinides, including ²³⁹Pu.⁴² In this case, the concentration ratio is expressed as the concentration of the nuclide in the plant (pCi/g) divided by the concentration in the soil (pCi/m³). The range of concentration ratios was from 10⁻⁶ to 10⁻¹¹. In this study, which included measurements of concentration ratios in barley, peas, beans, soybeans, wheat, and tomatoes, the ²³⁹Pu concentration in the edible part of the plant (e.g., wheat grain) was much smaller than that in the nonedible part of the plant (e.g., wheat straw).

These results are consistent with a laboratory study conducted in the United Kingdom by scientists at the National Radiological Protection Board.⁴³ In this study, carrot, cabbage, and barley crops were grown in loam, peat, and sandy soils, artificially contaminated with various radionuclides. The concentration ratios of the various radionuclides were measured for edible and nonedible parts of the

crops as a function of soil type, crop, and radionuclide. For all crops and all soil types, the measured concentration ratios for $^{239,240}\text{Pu}$ were less than 0.001. Additionally, the concentration ratios in the edible parts of the crops were lower than those in the nonedible parts.

A similar study was conducted by scientists at the University of California at Los Angeles, who measured concentration ratios of radioactive actinides in peas, soybeans, tomatoes, and wheat grown in seven different soil types.⁴⁴ The soils were selected to represent those associated with the major food producing areas of the United States. Of the actinides included in this study, the concentration ratios for plutonium were the smallest, with values associated with the edible parts of the crops ranging from 10^{-4} to 10^{-6} .

As previously stated, once plutonium has been adsorbed onto soil constituents, it tends to become relatively immobile and to migrate only very slowly with the groundwater. However, it is possible to make it more soluble by the addition of chelating agents. These are chemicals that complex with the plutonium compounds, changing their chemical behavior. When these agents are added to the soil, the plutonium compounds become much more mobile and can then be extracted from the soil, either through intentional remediation or via uptake to plants. Scientists at the Bhabha Atomic Research Center in Bombay, India, measured the effects of two chelating agents on the uptake of plutonium by plants in two soil types.⁴⁵ In this study, two chelating agents were used, DTPA and EDTA, in two soil types and the uptake by beans increased by factors ranging from 3 to 700 over the uptake in soils not treated with these chelating agents.

Plutonium in Air

Plutonium can become entrained in air through several mechanisms. First, when a nuclear weapon is exploded, especially if it is a ^{239}Pu -based device, a significant mass of this nuclide is forced high into the atmosphere, where it is distributed via upper level winds over much of the earth. Over time, the ^{239}Pu falls by gravity or is washed by precipitation from the air and is deposited onto the ground, a phenomena known as fallout. Since the last atmospheric nuclear explosions occurred several decades ago, essentially all of the fallout ^{239}Pu has been removed from the atmosphere. The second mechanism is if a nuclear reactor sustains a severe core-damage accident, wherein a significant fraction of the core's inventory of ^{239}Pu is vaporized and can, potentially, be injected into the atmosphere by the thermal energy generated during the accident. An example of this type occurred in the Chernobyl reactor accident, which took place over a 10-day period starting on April 26, 1986. Measurements of air concentrations of $^{239,240}\text{Pu}$ in Belgrade, Yugoslavia were made during the first two weeks of May 1986 to determine the plutonium fallout from the accident.⁴⁶ Air concentrations ranged from 0.4 to $10.6\ \mu\text{Bq}/\text{m}^3$. Again, scrubbing of the air, either by gravity or by precipitation, has largely removed all of the ^{239}Pu that was released during this accident.

In areas far removed from the Chernobyl area, the dominant source of plutonium contamination is fallout from weapons testing. Enough time has passed since the last atmospheric test in 1980 to allow measurement of baseline air concentrations of the plutonium isotopes.⁴⁷ This baseline concentration at the Pacific Northwest National Laboratory, near Richland, Washington, is $0.13\ \mu\text{Bq}/\text{m}^3$. The baseline concentration for the Argonne National laboratory - East, near Chicago, Illinois, ranges from 0.06 to $0.14\ \mu\text{Bq}/\text{m}^3$.

The third mechanism by which ^{239}Pu can end up in air is by being entrained by wind in air (as an aerosol) and transported before eventually falling back to earth's surface. This can happen either over land (with contaminated soil) or a sea (with contaminated water). The source of contamination can be fallout or discharge of fluids from a nuclear facility (e.g., nuclear reprocessing facility). Scientists from the Harwell Laboratory conducted a study of the transfer of radionuclides (including $^{239,240}\text{Pu}$) from

offshore water to land via wind-induced entrainment.⁴⁸ As radionuclides are discharged into seawater, they partition between two phases in the water: a soluble phase, wherein the radionuclide dissolves into the water; and a particulate phase, wherein the radionuclide is adsorbed onto a small soil particle, which is suspended in the water. Then, agitation of the water surface, due to wind shear, causes either phase to become airborne and the wind subsequently transports the radionuclide onto the land. The relative distribution between soluble and particulate-associated phases is nuclide dependent. For the case of ²³⁹Pu, the soluble phase is relatively insignificant and the transport is dominated by the particulate phase—approximately 95% of the water concentration of this nuclide is associated with particulates. Soil contamination due to this mechanism is very dependent on the distance from the shoreline. Soil contamination near the Dounreay Nuclear Power Development Establishment (northern Scotland) due to contaminated aerosols from the Pentland Firth indicate surface contamination of ^{239,240}Pu ranging from more than 500 Bq/m² near the shoreline to about 50 Bq/m² 1/2 km inland.

An example of the contamination of air with ^{239,240}Pu due to wind entrainment of contaminated soil was documented by measurements taken near the nuclear weapons testing sites of Maralinga and Emu in Australia.⁴⁹ High Volume Air Samplers were used to collect atmospheric samples, which were analyzed for radionuclide concentration. Most of the time, the airborne concentrations of plutonium were less than 10⁻⁴ Bq/m³. However, occasional higher concentrations were measured (average [over a few days] of 10⁻² Bq/m³ and higher) during periods of high wind velocities.

Resuspension of ^{239,240}Pu from topsoil in the Czech Republic has been measured by scientists at the National Radiation Protection Institute.⁵⁰ The soil was contaminated with the nuclide due to fallout from the accident at the Chernobyl reactor. Over a period of three years of measurements, the range of ^{239,240}Pu concentration in the air of Prague ranged from <0.5 to 5.5 nBq/m³.

The area surrounding the Chernobyl reactor complex received, of course, the highest soil contamination levels from the 1986 reactor accident. Soil concentrations of ^{239,240}Pu in the region within 30 km of the reactor range from 700 to 7,000 Bq/kg.⁵¹ Resuspension of plutonium from the soil, more than 1 year after the accident, resulted in air concentrations up to 100 µBq/m³ in the city of Chernobyl (approximately 15 miles from the reactor complex) and up to 380 µBq/m³ in Prip'yat, a town approximately 1/2 mile away from the reactor.

BEHAVIOR OF PLUTONIUM IN THE HUMAN BODY AND IN ANIMALS

The behavior of ^{239}Pu in the human body is generally based on two sets of data. First, data have been collected on the health effects in humans exposed to this nuclide, either as a result of nuclear explosions (e.g., survivors of the Nagasaki nuclear bomb explosion in August 1945) or due to working in jobs involving plutonium (e.g., workers involved in nuclear weapons research and production). The second database has resulted from studies of the metabolism and radiological effects of ^{239}Pu in animals—these results are then extrapolated to humans. The first database is of more value since there is less need to extrapolate the results to humans. However, these data are relatively sparse since they are principally dependent on accidental exposures. Thus, data from animal studies play an important role in understanding the behavior of this nuclide in humans.

The principal radiological hazard associated with ^{239}Pu is due to its high-energy, α -particle emissions (average α -particle energy is 5.15 MeV). The beta and gamma emissions are relatively low energy and are not as radiologically significant. Since even these high-energy α particles cannot penetrate the skin, external exposure to ^{239}Pu is much less of a hazard than internal exposure. Therefore, to pose a significant health hazard, this nuclide must enter the human body. There are several ways in which this may occur. First, plutonium contamination of a wound (cut or abrasion) can result in absorption into the blood and transport to other organs. Second, plutonium may be ingested by eating contaminated food. Third, plutonium may be injected into either muscle tissue or the blood stream (this means is used in experiments and does not pose a significant threat to the general population). Fourth, contaminated air can be inhaled, contaminating the lungs and other body organs. Of these contamination methods, ingestion is considered to be relatively benign. This is because plutonium is poorly absorbed across the gastrointestinal tract.⁵² In fact, most of the transuranic compounds are not readily absorbed from the gastrointestinal tract. Of these, however, plutonium is one of the least absorbed—for example, for PuO_2 , only 0.0001% of the ingested compound is absorbed in rats.⁵³

Reference 54 further states that inhaled PuO_2 is largely retained in the lungs and thoracic lymph nodes and that absorption through the skin is relatively insignificant. Retention in the lung is significantly affected by the physical properties of the inhaled particles, especially particle size. Smaller particles tend to more easily translocate. From the lungs, the plutonium compounds are translocated to other organs, with the distribution dependent on the compound. For example plutonium citrate and nitrate tend to locate to bone and, to a lesser extent, to the liver. This observation is consistent with a German study, which concluded that the majority of plutonium, originating from fallout, is concentrated in the skeleton and liver organs of animals.⁵⁴

The effects of inhaled PuO_2 in rats were measured in a study conducted by scientists from Pacific Northwest National Laboratory.⁵⁵ It was shown that the preparation of the oxide was significant in the toxicity of the compound. Some aerosols were used that had been oxidized in air at room temperature. In addition, some plutonium was oxidized at high temperatures, and the effects of the difference in preparation were measured. The air-oxidized aerosols were much more toxic, as revealed by the number of rats with induced lung tumors, than those that were oxidized at high temperature.

Biological half-life is a measure of the length of time that plutonium compounds are retained in the body. This half-life is strongly influenced by the specific compound. For example, organic complexes of plutonium, plutonium nitrate, and plutonium fluoride have biological half-lives that range from 30 to 300 days in rats, hamsters, and dogs. The half-lives for plutonium oxide range from 150 to 500 days in rodents and 300 to 1,240 days in dogs.⁵⁴

Because of the very long half-life of ^{239}Pu , its retention in the human body is dictated by biological processes instead of radiological decay. A biological half-life is defined as the length of time half of the

mass of ^{239}Pu associated with a specific body part is eliminated from the body due to biological processes (such as excretion or urination).⁵⁶ The biological half-life for plutonium depends on the specific body part in question. This reference lists the following biological half-lives in humans for plutonium: 65,000 days (178 y) for the whole body; 73,000 d (200 y) for bones; and 30,000 d (82 y) for the liver. Thus, once this nuclide is absorbed into the human body, it will tend to stay, with only minor removal, as long as the person is alive.

The distribution of ^{239}Pu in human bodies is one of the focuses of the U.S. Transuranium Registry (USTR), the only research program in the U.S. that is actively studying the internal depositions of Pu and the higher actinides in human tissues.⁵⁷ This agency performs radiochemical analysis and evaluation of selected tissues and organs as well as of the whole body of people who have been exposed to transuranic elements (usually through work activities) and who voluntarily donate their bodies to the program. As of December 1997, there were a total of 889 individuals who have volunteered to participate in the program. Of these, 359 have died, 266 are living and still participate in their work activities, and 264 are living but retired. These represent a vast wealth of actual and potential data that will greatly assist epidemiologists in assessing the potential health effects of exposure to plutonium and the other transuranic elements.

Reference 58 contains the results of the examination of a Caucasian male who died at age 62 of respiratory failure associated with pneumonia. This man had been employed from January 1945 to February 1982 at a site where he was involved in operations that potentially resulted in exposure to plutonium. This was especially true of the early years of his employment, when worker protection measures were much less sophisticated than today. A crude estimate of his exposure was made based on historical radiourinalyses and nasal wipes. It is estimated that he inhaled as much as 1,000 Bq of ^{239}Pu , most of it during the first 1-1/2 years of his employment. The early exposure was to a mixture of plutonium fluoride and oxide and the late exposures, to oxides of plutonium. Assuming that the total dose estimate of 1 kBq is correct, approximately 75% of the inhaled ^{239}Pu had been removed from the body prior to death. Approximately one-half of the total retained ^{239}Pu remained in the respiratory tract, based on the autopsy performed by the USTR. The rest of the retained plutonium was approximately equally distributed between the liver and the skeleton, with minor depositions in other organs.

In another USTR study, autopsies were performed on two former nuclear workers.⁵⁸ The first one had worked as a research chemist from 1950 to 1979, when he died from a metastatic malignant melanoma. The second one was employed from 1945 to 1980 as a chemical engineer in plutonium alloy research and development and in plutonium weapons work. He had a history of heart problems and died, one year after retirement, from heart failure compounded by pneumonia. In both cases, it is judged that exposure to ^{239}Pu occurred due to inhalation of contaminated air. The results from both examinations indicate that approximately 45% of the systemic (whole body less the respiratory tract) plutonium burden is in the liver and 45% in the skeleton, with the remaining 10% in other tissues.

The International Commission on Radiological Protection (ICRP) performed a survey of the available data regarding the distribution of plutonium in the human body.⁵⁹ Table 6.3 of this reference lists the results of several studies involving occupational exposure to plutonium. While the total relative (percent) inventory associated with the liver and skeleton are consistent among the individual studies (representing approximately 75–80 % of the total deposited plutonium), the distribution between these two organs differs significantly. In some of the studies, there was approximately twice as much plutonium in the liver as in the skeleton and in others, the ratio is reversed. The authors indicate that this may be due to redistribution within the body as a person ages. For example, there is some evidence that as a person ages, plutonium may migrate from the skeleton to the liver, although these results are not conclusive.

Table 7 lists limits recommended by the ICRP for those who work in environments where they might be exposed to ^{239}Pu , either by inhalation or ingestion. Two quantities are included in the table. First, the Annual Limits on Intake or ALI, which limits the total amount of ^{239}Pu that should be incorporated into a person over one year's time. Second, the Derived Air Concentrations (DAC) limits the air concentration of ^{239}Pu for a worker who is in the environment for 40 hours per week. An inhalation classification is assigned, depending on whether the specific chemical compound tends to be cleared from the lungs in days (D), weeks (W), or years (Y). For plutonium, none of the chemical compounds are classified D. Oxides are classified Y and all other commonly occurring compounds are classified W. The f_1 value represents the fraction of the inhaled or ingested plutonium that is transferred to the circulatory system and is also associated with the classification.

Table 17-7. Annual limits on intake (ALI) and the derived air concentrations (DAC) for ^{239}Pu for ingestion and inhalation. ⁶⁰

Species	Component	Ingestion per year and (f_1 value) ^a	Inhalation and (/Class and f_1 value) ^a
^{239}Pu -oxides	ALI	$3 \times 10^6 \text{ Bq } (f_1 = 10^{-5})$	600 Bq /Y ($f_1 = 10^{-5}$)
^{239}Pu -all others		$3 \times 10^4 \text{ Bq } (f_1 = 10^{-3})$	200Bq/W ($f_1 = 10^{-3}$)
^{239}Pu -oxides	DAC	— ^b	0.3 Bq/m ³ /Y ($f_1 = 10^{-5}$)
^{239}Pu -all others			0.1 Bq/m ³ /W ($f_1 = 10^{-3}$)

a. Values depend upon the removal time of the specific chemical form of ^{239}Pu and are placed in the three possible lung clearance classes: D representing days, W for weeks, or Y for years. f_1 represents the fraction of the substance that enters the blood-stream via the gastrointestinal tract or from the lung with a clearance class of D, W, or Y. For ingestion calculations, plutonium oxides have a $f_1=10^{-5}$ with a classification of Y and all other common plutonium compounds have a $f_1=10^{-3}$ with a classification of W.

b. Data not available.

Plutonium has gained a reputation as the most feared substance on earth. It has been characterized as the most toxic substance known to man and is believed, by many, to be so deadly that uptake in the body of even the slightest amount (sometimes termed a “speck”) will guarantee the person develops terminal cancer. While plutonium is a very toxic substance and represents a definite radiological hazard, if not handled correctly, it is far from being the most toxic substance known to man. In fact, there are several substances that are many times more toxic than plutonium, as measured by their acute toxicity, or the specific amount that results in death.

Toxicity can be split into two components, acute toxicity and latent toxicity. Acute toxicity is the ability of a toxin to cause death shortly after exposure. Latent toxicity is the ability of a toxin to cause death long times after exposure. In the case of ^{239}Pu , the acute toxicity is due to the chemical effects of this element (or compound, depending on the exposure), whereas latent toxicity is due to its radiological characteristics.

Since any toxin reacts somewhat differently in different people or other organisms, the study of toxicity requires a statistical approach. Accordingly, a measure of acute toxicity is LD_{50} . This term is defined as the amount of toxin (usually measured in mass of toxin divided by the mass of the organism) that results in the short-term death of 50% of a large number of similarly exposed organisms. Thus, for example, if the LD_{50} for a given toxin is 1 g/kg, this means that incorporation of 1 g of the toxin per kg of organism mass in a population of 1,000 will result in the near-term death of 500. The acute LD_{50} of plutonium in various mammals, such as dogs, rats and mice, is approximately 1 mg/kg.⁶¹ Extrapolated to man, it would take exposure to 0.1 g of plutonium to kill 50% of a population of 100-kg men. Other

toxins are much more lethal. Strychnine, a common poison, has an LD₅₀ of 500 µg/kg, which means it is twice as toxic as plutonium. Curare, commonly used as a muscle relaxant during surgery, has a similar LD₅₀. Diphtheria toxin has an LD₅₀ of 1×10^{-4} µg/kg, which means that it is 10 million times more toxic than plutonium. Crystalline botulinus toxin has an LD₅₀ of 7×10^{-9} µg/kg, which means it is 14 billion times more toxic than plutonium. All of these values are for the short-term or acute toxic effects.

The latent effect of exposure to ²³⁹Pu was the subject of a Los Alamos National Laboratory study.⁶² This paper provides an update to an ongoing study of the health of a group of 26 young males who worked on the Manhattan Engineer District's Project "Y" at Los Alamos in 1944 and 1945. During this time, these men experienced significant contamination of ²³⁹Pu. Of the 26, 21 had left Los Alamos by 1946, one left in 1948, and none had significant additional plutonium exposure after 1945. Exposure was primarily by inhalation, though some also were exposed through cuts in the skin. The internal plutonium deposition ranged from 50 to 3,180 Bq and the effective dose ranged from 0.1 to 7.2 Sv. The median deposition and dose were 565 Bq and 1.25 Sv, respectively. The mortality of this group was compared to the mortality of U.S. white males. The standardized mortality ratio (STR), calculated by comparing the mortality of the subject group to the U.S. white male population, indicates that the exposed Los Alamos group experienced statistically significant fewer cancers and longer lives. Since this may be due to the education level of the Los Alamos group (these 26 had attained a higher level of education than the average U.S. male and this generally correlates with a healthier life style), they were also compared with a population of unexposed Los Alamos workers with comparable hire dates and general education levels. This comparison indicates that the general mortality, as well as cancer-induced mortality, of the two groups were statistically similar. Thus, the exposure of these 26 men to ²³⁹Pu did not significantly affect their health over their lifetimes.

A similar study was conducted by scientists associated with the Russian Ministry for Public Health and Medicinal Industry.⁶³ In this study, the incidence of lung cancer among a group of 500 nuclear workers was investigated. Of the 500, 162 contracted lung cancer and eleven potential risk factors were evaluated using a logistic regression model. This was done to determine the relative contribution each of these factors made to the incidence of cancer. The workers were exposed to airborne ²³⁹Pu in a range from 0–141 kBq. Five of the factors were determined to have not contributed significantly to the cancer incidence. The remaining six factors were arranged in decreasing order of significance. The odds ratio (OR), a parameter that measures the relative contribution of the various factors to the incidence of cancer, was determined for each of these six factors. The most significant contributor to lung cancer, among these workers, was smoking, with an OR of 6.6. The OR for plutonium incorporation was 1.3 for the whole professional contingent and 3.1 for those professionals who incorporated more than 5.55 kBq. These OR values (for plutonium incorporation) are similar in magnitude to those for workers who inhale other harmful substances—metal workers, 2.4; asbestos workers, 1.7; chimney-sweeps, 2.1; rubber production workers, 3.2.

In summary, ²³⁹Pu is a hazardous material and can result in severe health effects if a person is contaminated with it. However, if handled carefully, these health effects can be avoided. While toxic, it is certainly not in the category of "super toxins," such as certain disease strains and industrial poisons.

SUMMARY

^{239}Pu is an artificially produced radionuclide that does not normally exist in nature. Its half-life is approximately 24,100 years and it decays by α -particle emission to ^{235}U . When it decays, α -particles of maximum energy of 5.156 MeV are released. In addition, atomic electrons and γ -rays (from ^{235}U) are almost simultaneously released. The average energy of the released α -particles is 5.15 MeV. There are no high energy γ or β emissions associated with this nuclide and, therefore, it does not pose a significant external radiological hazard. However, this nuclide is classified in the very high radiological hazard group because of its high-energy α -particle, which can cause significant biological damage if the nuclide is incorporated into the body.

^{239}Pu is primarily produced in nuclear reactors (for example in a fast breeder reactor) through neutron absorption of ^{238}U , which produces ^{239}U . This nuclide decays via β -decay to ^{239}Np with a half-life of 24 minutes. ^{239}Np , in turn, decays (also by β -decay) to ^{239}Pu with a half-life of 24,100 years. Thus, in a very short time (of the order of a few weeks) all of the ^{239}U has decayed to ^{239}Pu .

The principal uses of this nuclide are associated with its fissile nature. Since it can sustain a chain reaction, it can be used as fuel in a nuclear reactor. For this use, it is usually formed into an oxide and is often mixed with UO_2 to form a mixed oxide nuclear fuel. The other principal use of this nuclide is as a weapons material in nuclear fission and fusion bombs.

The chemistry of this element is complex in that it can form hundreds of different compounds. In the absence of surface oxidation, plutonium is a silvery-white metal, similar to nickel. The better-known plutonium compounds that are important to the nuclear industry and fuel fabricators are plutonium-oxides, plutonium-hydrides, plutonium-fluorides, plutonium-chlorides, plutonium-carbides, plutonium-nitrides, and plutonium-sulfides, as well as many other compounds.

When plutonium is deposited onto soils (for example, through fallout or as a result of discharges from a plutonium facility), it tends to be readily retained in the soil, usually within the top few centimeters of the soil. Also, it is not readily absorbed via the roots into plants and its presence in vegetation usually occurs through air contamination. When it is absorbed by the roots, it tends to concentrate in the nonedible parts of the plant.

The majority of data concerning plutonium effects on man are based on extrapolation of laboratory studies of small animals; however, a small number of humans injected with plutonium or accidentally exposed with plutonium have been studied. Plutonium is generally difficult to absorb from the gastrointestinal tract and the most hazardous means of internal contamination is via inhalation. When it does enter the circulatory system (either through ingestion and subsequent absorption in the gut or through inhalation and absorption in the lungs), it tends to be concentrated in either the liver or the skeletal system. Once it is deposited in either the liver or skeletal system, it tends to remain in the human body for a very long time, usually as long as the person is alive.

^{239}Pu has gained a reputation as “the most toxic substance known to man.” This reputation is undeserved as there are several substances that are more toxic than ^{239}Pu in the short term. In the long term, there is no question that this nuclide is hazardous and can cause bodily harm if incorporated into the body. However, workers in the plutonium industry have not experienced significantly higher cancer incidence, even though their exposure in many cases exceeded current standards for radiation workers.

REFERENCES

1. General Electric Company, *Nuclides and Isotopes*, Fifteenth Edition, Revised 1996, p. 48.
2. M. M. Benjamin, *Nuclear Reactor Materials and Applications*, Van Nostrand Reinhold Company, 1983, p. 208, and Chapter 8.
3. Isotope Distribution Office of Oak Ridge National Laboratory, "Radioisotope Price List," Revised October 1, 1993, Phone: (423) 574-6984. Also see the following Internet address: [<http://www.ornl.gov/isotopes/catalog.htm>] for additional isotope information.
4. F. J. Rahn et al., *A Guide to Nuclear Power Technology*, New York: John Wiley & Sons, 1984, Chapter 5.3 pp. 99, 171–181.
5. F. A. Mettler, Jr., M.D., A. C. Upton M.D., *Medical Effects of Ionizing Radiation*, Second Edition, W. B. Saunders Company, 1995, pp. 156 and 346–349.
6. B. G. Bennett, "Transfer of Plutonium from the Environment to Man," *Proceedings of the International Symposium on the Management of Wastes from the LWR Fuel Cycle*, July 11–16, 1979, Denver, Colorado, Sponsored by the Energy Research and Development Administration, and arranged by Oak Ridge National Laboratory, CONF-76-0701, July 1976, pp. 88–90 and 554–564.
7. R. Granier and D. Gambini, *Applied Radiobiology and Radiation Protection*, Ellis Horwood Limited, English edition published in 1990, p. 283, and Appendix 2.
8. P. K. Park, D. R. Kester, I. W. Duedall, and B. H. Ketchum, *Wastes in the Ocean*, Vol. 3, New York: John Wiley & Sons, 1983, pp. 8, 9, 22, 26, 28.
9. Z. A. Medvedev, *The Legacy of Chernobyl*, W. W. Norton & Company, 1990, pp. 78 (Table 3.1), 98, and 99.
10. W. C. Hanson (Editor), *Transuranic Elements in the Environment*, Published by the Technical Information Center/U.S. Department of Energy, 1980, pp. 54, 59, 88, 89, 90, 96, 107.
11. O. J. Wick (Editor), *Plutonium Handbook A Guide to the Technology*, Volume I, Gordon and Breach, Science Publishers, 1967, pp. 8, 13, 27, 147, Chapters 11 and 12.
12. Nuclear Data Retrieval Program from the National Nuclear Data Center, Brookhaven National Laboratory, Upton, NY 11973. See Internet addresses: <http://www.nndc.bnl.gov> or <http://www.nndc.bnl.gov/nndc/nudat/radform.html>.
13. C. M. Lederer, J. M. Hollander, I. Perlman, *Table of Isotopes*, Sixth Edition, New York: John Wiley and Sons, Inc., 1967, p. 144 for ²³⁹Pu, ⁴²⁷ and ⁴³².
14. Henry Semat, *Introduction to Atomic and Nuclear Physics*, Fourth Edition, Published by Holt, Rinehart and Winston 1963, p. 345 and 359.

15. U.S. Department of Health, Education, and Welfare, *Radiological Health Handbook*, Revised Edition, January 1970, pp. 122 and 204.
16. Microshield dose calculation: [C. A. Negin, G. Worku, *MicroShield, Version 4.10, User's Manual*, Grove Engineering, Inc., 15215 Shady Grove Road, Rockville, Maryland 20850.
17. R. L. Murray, *Understanding Radioactive Waste*, Third Edition, Published Battelle Press, 1989, p. 21.
18. A. R. Kaufmann, *Nuclear Reactor Fuel Elements Metallurgy and Fabrication*, Interscience Publishers, 1962, p. 105 (Table 4-3), or p. 115.
19. D. C. Stewart, *Data for Radioactive Waste Management and Nuclear Application*, Published by John Wiley & Sons, 1985, p. 263.
20. R. C. Weast (ed.), *CRC Handbook of Chemistry and Physics*, CRC Press, 1977–1978, 58th Edition, pp. B270-B354 (Table of Isotopes), pp. B-141 and B-142.
21. A. G. Croff, *A User's Manual for the ORIGEN2 Computer Code*, ORNL/TM-7175, July 1980.
22. Oak Ridge National Laboratory, U.S. Department of Energy, Office of Environmental Management, *Integrated Data Base Report – 1996: U.S. Spent Nuclear Fuel and Radioactive Waste Inventories, Projections, and Characteristics*, DOE/RW-006, Rev. 13, December 1997.
23. P. J. Coughtrey et al., *Radionuclide Distribution and Transport in Terrestrial and Aquatic Ecosystems: A Critical Review of Data*, Volume Four, A. A. Balkema, 1984, pp. 1, 2, 5, 7, 8, 9, 19, 20, 21, 26, 27, 34, 118, 224 (Chapter 29).
24. T. M. Poston, E. J. Antonio, and A. T. Cooper, *Radionuclide concentrations in Terrestrial Vegetation and Soil on and Around the Hanford Site, 1983 through 1993*, PNL-10728, UC-602, August 1995.
25. C. S. Kim et al., “ ^{90}Sr , ^{137}Cs , $^{239+240}\text{Pu}$, and ^{238}Pu Concentrations in Surface Soils of Korea,” *Journal of Environmental Radioactivity*, Vol. 40, p. 75, 1998.
26. H. Nishita, et al., “Effect of Soil Type on the Extractability of ^{237}Np , ^{239}Pu , ^{241}Am , and ^{244}Cm as a Function of pH,” *Soil Science*, Vol. 132, July 1981, p. 25.
27. W. C. Hanson, Ed., *Transuranic Elements in the Environment*, DOE/TIC-22800, p. 29, 1980.
28. Y. Mahara and H. Matsuzuru, “Mobile and Immobile Plutonium in a Groundwater Environment,” *Water Resources*, Vol. 23, p. 43, 1989.
29. K. Bunzl, W. Kracke, and W. Schimmack, “Migration of Fallout $^{239+240}\text{Pu}$, ^{241}Am , and ^{137}Cs in the Various Horizons of a Forest Soil Under Pine,” *Journal of Environmental Radioactivity*, Vol. 28, p. 17, 1995.

30. V. A. Knatko, et al., "Characteristics of ^{90}Sr , ^{137}Cs , and $^{239,240}\text{Pu}$ Migration in Undisturbed Soils of Southern Belarus after the Chernobyl Accident," *Journal of Environmental Radioactivity*, Vol. 30, p. 185, 1996.
31. H. Nishita and M. Hamilton, *Soil Components that Influence the Chemical Behavior of ^{239}Pu* , UCLA-12-1159/XAB, August 1978.
32. D. G. Korich and J. E. Sharp, *Bioreduction Amenability Testing of Actinide Contaminated Soils*, LA-SUB-95-208, January 1995.
33. H. Nishita and R. M. Haug, "The Effect of Fulvic and Humic Acids and Inorganic Phase of Soil on the Sorption and Extractability of $^{239}\text{Pu(IV)}$," *Soil Science*, Vol. 128, pp. 291, 1979.
34. L. J. Kirby, Ed, *Radionuclide Distributions and Migration Mechanisms at Shallow Land Burial Sites*, NUREG/CR-3607, PNL-4823, February 1984.
35. D. M. Nelson et al., *Actinide Speciation in the Shallow Aquifer of Mortandad Canyon*, LA-9816-PR, August 1983.
36. D. E. Robertson, K. H. Abel, and B. E. Vaughan, *Transuranic Chemical Species in Ground Waters*, PNL-41000-Pt. 2, p. 81, February 1982.
37. C. E. Cowan, et al., *Transuranic Chemical Species in Groundwater: Final Report*, PNL-5263, February 1985.
38. H. Nitsche et al., *Measured Solubilities and Speciations of Neptunium, Plutonium, and Americium in a Typical Groundwater (J-13) from the Yucca Mountain Region*. LA-12562-MS, July 1993.
39. A. B. Kersting, et al., "Migration of Plutonium in Ground Water at the Nevada Test Site," *Nature*, Vol. 397, pp. 56, 1999.
40. B. D. Honeyman, "Colloidal Culprits in Contamination," *Nature*, Vol. 397, pp. 23, 1999.
41. C. Murphy, *The concentration of Radionuclides and Metals in Vegetation Adjacent to and in the SRL Seepage Basins*, WSRC-TR-92-583, December 1992.
42. C. Murphy and R. Tuckfield, *Vegetation Uptake from Burial Ground Alpha Waste Trenches*, WSRC-RP-89-1916, 1989.
43. A. Nisbet and S. Shaw, "Summary of a Five-Year Lysimeter Study on the Time Dependent Transfer of ^{137}Cs , ^{90}Sr , $^{239,240}\text{Pu}$, and ^{241}Am to Crops from Three Contrasting Soil Types. 2 Distribution between Different Plant Parts," *Journal of Environmental Radioactivity*, Vol. 23, p. 171, 1994.
44. A. E. Romney et al., "Plant Uptake of ^{237}Np , $^{239,240}\text{Pu}$, ^{241}Am , and ^{242}Cm From Soils Representing Major Food Production Areas of the United States," *Soil Science*, Vol. 132, pp. 40, 1981.
45. B. N. Vyas and K. B. Mistry, "Influence of Chelating Agents on the Uptake of ^{239}Pu and ^{241}Am by Plants," *Plant and Soil*, Vol. 73, pp. 345, 1983.

46. S. Mani-Kudra et al., "Plutonium Isotopes in the Surface Air at Vinca-Belgrade Site in May 1986," *Journal of Radioanalytical and Nuclear Chemistry Letters*, Vol. 199, p. 27, 1995.
47. V. Pan and K. Stevenson, "Temporal Variation Analysis of Plutonium Baseline Concentration in Surface Air from Selected Sites in the Continental US," *Journal of Environmental Radioactivity*, Vol. 32, p. 239, 1996.
48. W. McKay and N. Pattenden, "The Transfer of Radionuclides from Sea to Land via the Air: A Review," *Journal of Environmental Radioactivity*, Vol. 12, p. 49, 1990.
49. P. Johnston, et al., *Inhalation Hazard Assessment at Maralinga and Emu, Chapter 2: Ambient Concentrations of Radionuclides in Air at Maralinga and Emu*, ARL-TR-087, May 1990.
50. Z. Hölgge and R. Filgas, "Concentrations of $^{239,240}\text{Pu}$ and ^{238}Pu in the Surface Air of Prague in 1993, 1994, and 1995," *Journal of Radioanalytical and Nuclear Chemistry Letters*, Vol. 214, p. 303, 1996.
51. Y. Gaziev, et al., "Procedures and findings of Examination of Air Contamination by Alpha-Active Plutonium Aerosols: Primary and Secondary Products of Chernobyl Accident," *Journal of Radioanalytical and Nuclear Chemistry*, Vol. 150, p. 201, 1991.
52. P. Coughtrey, D. Jackson, and M. Thorne, *Radionuclide Distribution and Transport in Terrestrial and Aquatic Ecosystems - A Compendium of Data*, A. A. Balkema, 1984, Vol. 6, p. 117.
53. W. Bair, "Recent Animal Studies on the Deposition, Retention and Translocation of Plutonium and Other Transuranic Compounds," *IAEA International Seminar on Diagnosis and Treatment of Incorporated Radionuclides*, Vienna, Austria, December 8, 1975, p. 44.
54. K. Bunzl and W. Krancke, "Fallout $^{239,240}\text{Pu}$ and ^{137}Cs in Animal Livers Consumed by Man," *Health Physics*, Vol. 46, p. 466, 1984.
55. C. Sanders and J. Mahaffey, "Carcinogenicity of Inhaled Air-Oxidized $^{239}\text{PuO}_2$ in Rats," *International Journal of Radiation Biology*, Vol. 35, p. 95, 1979.
56. Wang, Y., (ed), *CRC Handbook of Radioactive Nuclides*, Boca Raton, Florida: CRC Press, 1969.
57. J. McInroy, et al., "U.S. Transuranium Registry Report on the ^{239}Pu Distribution in a Human Body," *Health Physics*, Vol. 60, p. 307, 1991.
58. J. McInroy and M. Swint, "Distribution of ^{239}Pu and ^{241}Am in the Human Skeleton," *18th European Symposium on Calcified Tissue*, Angers, France, October 11, 1984, p. 14.
59. "The Metabolism of Plutonium and Related Elements," *Annals of the ICRP - ICRP Publication 48*, Vol. 16, No. 2/3, 1986.
60. "Limits for Intakes of Radionuclides by Workers: an Addendum," *Annals of the ICRP - ICRP Publication 30 Part 4*, Vol. 19, 1988.

61. J. Stannard, *Radioactivity and Health - A History*, DOE/RLI/01830-T59, October 1988.
62. G. Voelz, J. Lawrence, and E. Johnson, "Fifty Years of Plutonium Exposure to the Manhattan Project Plutonium Workers: An Update," *Health Physics*, Vol. 73, p. 611, 1997.
63. Z. Tokarskaya, et al., "The Influence of Radiation and Nonradiation Factors on the Lung Cancer Incidence Among the Workers of the Nuclear Enterprise Mayak," *Health Physics*, Vol. 69, p. 356, 1995.

BIBLIOGRAPHY

- “Age-Dependent Doses to Members of the Public from Intake of Radionuclides: Part 4 Inhalation Dose Coefficients,” *Annals of the ICRP*, Vol. 25, No. 3 - 4, 1995.
- “Age-Dependent Doses to Members of the Public from Intake of Radionuclides: Part 5 Compilation of Ingestion and Inhalation Dose Coefficients,” *Annals of the ICRP*, Vol. 26, No 1, 1996.
- Arthur, W., “Radionuclide Concentrations in Vegetation at a Solid Radioactive Waste Disposal Area in Southeastern Idaho, USA,” *Journal of Environmental Quality*, Vol. 11, No 3, p. 394, 1982.
- Ballou, J. E., et al., “Influence of DTPA on the Biological Availability of Transuranics,” *Health Physics*, Vol. 34, pp. 445, 1978.
- Cawse, P. and C. Colle, “Comparison of Radionuclide Deposition to Soil and Vegetation,” *Cadarache International Symposium on Radioecology*, Cadarache, France, pp. D75, March 14, 1988.
- Cawse P, et al., “Accumulation of Caesium-137 and Plutonium-239 + 240 in Soils of Great Britain and Transfer to Vegetation,” *Industrial Ecology Group of the British Ecological Society Meeting*, Essex, UK, pp. 47, April 5, 1982.
- Champ, D., et al., “Long-Lived Radionuclide Transport - An In-Situ Field Column Study,” *Transport and Mass Exchange Processes in Sand and Gravel Aquifers: Field and Modeling Studies Conference and Workshop*, Ottawa, Canada, pp. 725, October 1 - 4, 1990.
- “Dose Coefficients for Intakes of Radionuclides by Workers,” *Annals of the ICRP*, ICRP Publication 68, Vol. 24, No. 4, 1994.
- Duursma, E., “Plutonium in the Rhine-Meuse-Scheldt Delta: Analysis of Estuarine Sediment, Salt-Marsh Soil and Vegetation Samples,” *Radiation Protection*, pp. 285, Harwood Academic Publishers, NY, NY, 1981.
- Gvozdev, A., et al., “Radioecological Monitoring of Groundwater in the Areas of Storage Sites for Radioactive Waste Generated During Decontamination Work,” *International Symposium on Experience in the Planning and Operation of Low Level Waste Disposal Facilities*, Vienna, Austria, pp. 533, June 17 - 21, 1996.
- Kennedy, V. H. et al., “Use of Single and Sequential Chemical Extractants to Assess Radionuclide and Heavy Metal Availability from Soils for Root Uptake,” *Analyst*, Vol. 122, pp. 89, 1997.
- Kolb, W., “Remarks About the Seasonal Changes and Trends of the Pu-238, Pu-239+240, and Cs-137 Concentrations in Ground-Level Air,” *Proceedings of the Second International meeting on Low-Level Air Radioactivity Monitoring*, Madralin, Poland, pp. 45, February 14 - 18, 1994.
- McLeod, K. W. et al., “Uptake of Plutonium from Soils Contaminated by a Nuclear Fuel Chemical Separations Facility,” *Soil Science*, Vol. 132, pp. 89, 1981.

- Nisbet, F. and S. Shaw, "Effects of the Herbicide Glyphosate on the Uptake of ^{239}Pu and ^{241}Am to Vegetation," *Workshop on the Transfer of Radionuclides in Natural and Semi-Natural Environments*, Udine, Italy, September 11 – 15, 1989.
- Nisbet, a. And Shaw, S., "Effects of the Herbicide Blyphosate on the Uptake of ^{239}Pu and ^{241}Am to Vegetation," *Workshop on the Transfer of Radionuclides in Natural and Semi-Natural Environments*, Udine, Italy, pp. 554, September 11 - 15, 1989.
- Pan, V. And Stevenson, K., "Temporal Variation Analysis of Plutonium Baseline Concentration in Surface Air from Selected Sites in the Continental US," *Journal of Environmental Radioactivity*, Vol. 32, No. 3, pp. 239, 1996.
- Playford, K. and Baker, S., *Radioactivity in Air and Rainwater: UK Results to the End of 1996*, DETRA/RAS-97.004, AEAT--20069001/REMA.274, October 1997.
- Pulhani, V., et al., "Uptake of Radionuclides by Vegetation in the Natural Environment," *Bulletin of Radiation Protection*, Vol. 17, No. 1, pp. 36, Jan-Mar 1994.
- Rosner, G., Hoetzl, H., and Winkler, R., "Long-Term Behavior of Plutonium in Air and Deposition and the Role of Resuspension in a Semi-Rural Environment in Germany," *Science of the Total Environment*, Vol. 196 No. 3, pp. 255, March 5, 1997.
- Sanders, C. And Mahaffey, J., "Carcinogenicity of Inhaled Oxidized Plutonium-239 Dioxide in Rats," *International Journal of Radiation Biology Related Studies in Physics, Chemistry, and Medicine*, Vol. 35, No. 1, pp. 95, 1979.
- Swanson, J. L., "Organic Complexant-Enhanced Mobility of Toxic Elements in Low-Level Wastes," *6th Annual Low-Level Waste Management Program Participants' Information Meeting*, Denver, CO, September 11, 1984.
- Wallace, A. et al, "Role of Synthetic Chelating Agents in Trace Metal Uptake by Plants," *2nd Mineral Cycling Symposia on Environmental Chemistry and Cycling Processes*, Augusta, GA, April 28, 1976.